PRACTICAL APPLICATION OF IN SITU AEROSOL MEASUREMENT

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ABSTRACT

The use ***of in situ.*** real-time measurement techniques **permits** the characterization of airborne droplets and particles under conditions where traditional sampling methods can fail. **For** example, sampling methods rely on the ability to sample and transport particles without biasing the properties of interest. and ***often*** are not applicable in harsh environments. Although ***in situ*** methods offer unique opportunities in **these** cases. **these** techniques introduce new concerns and must be used carefully if accurate measurements are to be made. **There.** are numerous experimental **difficulties** inherent in spray droplet measurements: spatial nonuniformities: unsteady behavior: large spatial extent of the region of interest; **high** concentrations and wide concentration ranges; velocity distributions dependent **on** particle size: large variations in particle diameter (up to 3 orders ***of*** magnitude): evaporation, coalescence; fouling, deposition, and fogging of windows and optical surfaces: and **effects** of measuring probes ***on*** the droplets. For **these reasons. no** universal measuring device has yet been established. However, a number ***of*** droplet measurement techniques are available, based upon a variety ***of*** physical principles.

Several ***in situ*** measurement techniques are reviewed here.. As the field is rapidly evolving. **the** discussion is **limited to** those techniques which: 1) are commercially available. 2) provide real-time **output,** and 3) measure the **aerosol** size distribution. Discussion is divided between single panicle counters (which provide a flux-based or temporal measurement) and ensemble techniques (which provide a concentration-based or spatial measurement). Specific techniques discussed include phase Doppler. Mie scattering, and Fraunhofer diffraction, and commercial instruments based **on these** techniques.

INTRODUCTION

The characterization ***of*** airborne particles and droplets is critical in the study ***of*** a wide range of fields, including sprays, combustion. air quality, industrial processing. cleanroom monitoring, cloud and fog characterization. etc. The distributions ***of*** panicle size. shape. structure, charge. and chemical composition are each important in some context. In the broadest sense, the instruments provided for characterizing particles can be divided into two classes: extractive and ***in situ.*** In extractive sampling. a particle laden volume ***of*** **gas** is removed from its environment and transported to a separate. **location** where the particle measurement is made. Many of the most common **aerosol** measurement techniques operate in **this** mode. as it allows careful control of the conditions under which **the** measurement is made. The **success** of extractive techniques. however, relies **on** the ability to sample and transport particles without biasing the properties ***of*** **interest.** This condition is sometimes difficult to meet, **as** inlet inefficiencies. wall **losses,** and rapid **aerosol** dynamics (evaporation, condensation, coagulation) are examples of physical processes that can alter the particle size distribution Extractive techniques can also fail when measurements **need** to be made in hostile environments: extremes in pressure or temperature, reactive or corrosive environments, etc. ***In situ*** (noninvasive) measurement techniques can overcome many of these limitations, allowing particle characterization under conditions where extractive techniques are **not** suitable.

Several in *situ* measurement techniques are reviewed here. As the field is rapidly evolving, the discussion is limited to those techniques which: 1) are commercially available. 2) provide real-time output. and 3) measure the aerosol size distribution. Discussion is divided between single panicle counters (which provide a flux-based or temporal measurement) and ensemble techniques (which provide a concentration-based or spatial measurement). Among the aerosol measurement instrumentation in this review. capabilities exist for measurement of individual particle sizes from about 0.25 to above 1000 μm, concentrations as high as $10^6/cm^3$, and speeds in the kilometer/sec range. While *in situ* instruments overcome many of the limitations encountered with extractive methods. they do suffer (as a class and individually) from a wide range of new limitations. To describe these limitations. the next section provides an overview of in *situ* optical particle sizing systems, followed by a review of the instruments that are currently commercially available to the researcher. The individual reviews are by necessity short. but sufficient references are provided to help the reader to further explore each method. Although every effort has been made to include all of the available equipment, some manufacturers may have been overlooked.

OVERVIEW OF IN SITU INSTRUMENTS

The *in situ* measurement of particles by optical methods has been an area of active research - particularly over the last decade. Thus, many current reviews are available on the topic (Hirleman 1984.1988; Hovenac 1987: and Rader and O'Hern, 1993). Several recent sets of proceedings contain current applications and discussions of *in situ* techniques (Hirleman et al.. 1990; Hirleman, 1990; and Gouesbet and Grehan, 1988), and a recent issue of *Applied Optics* (*30*, 33.1991) was dedicated to papers on optical particle sizing.

It is helpful to divide optical in *situ* techniques into two general classes, based on whether they analyze single particle events a aggregate cloud properties. Single particle, counters (SPC) generally make a size determination on one particle at a time by analyzing its scattering behavior while it passes through a well defined (usually small) volume of high intensity (usually laser) light. Intensity, phase, or image information in the scattered light have all been used for particle sizing. A size distribution is obtained by sizing a number of particles sufficient to ensure statistical accuracy. SPC's generally provide a wealth of infamation on the *counted* panicles. providing correlations among partide properties such as size, velocity. and time of arrival. and allowing spatial characterization of the particle field. At high number concentrations, however. single partide counting techniques suffer from coincidence errors which occur when more than one particle occupies the sensing volume at the same instant.

The second class of in *situ* systems, collectively called ensemble techniques, generally operate by illuminating a volume containing a large number of particles and analyzing the collective scattering. An illustrative example of an ensemble technique would be a photographic snapshot (or a hologram in 3-D), which captures the state of a particle distribution at one instant in time. Ensemble techniques are well *suited* for measurements at high particle concentration. but become ineffective at low concentration. Generally, ensemble techniques do not provide as detailed information as SPC's. since individual particle information is lost in the averaging. Real-time ensemble techniques provide only limited spatial resolution of the particle field. Generally, ensemble techniques measure particle concentration (number/volume), while SPC systems measure particle flux (number/area/time) (Hirleman, 1988). That is. ensemble techniques report the number (and sizes) of particles present in the sampling volume over a short measurement time (spatial averaging), whereas SPC's report the number (and *sizes*) of particles passing through the sampling volume during a generally longer measurement time (temporal averaging). To obtain aerosol *concentration*, SPC's require additional particle velocity information. The distributions measured by concentration- or flux-based techniques will differ if a systematic correlation exists between partide *size* and velocity.

As each SPC or ensemble particle *sizing* technique offers distinct strengths and weaknesses. an ideal instrument can only be defined in terms of measuring a specific set of properties for a specific aerosol in a specific environment. In this vein. Hovenac (1987) and Hirleman (1988) outline an approach to *in situ* optical *sizing* in terms of instrument operating envelopes. The central idea is that the choice of instrument must be a two step process: first, *identify* the particle properties that need to be measured and the conditions under which the measurement must be made, and second, establish that these conditions fall within the instrument's operating envelope. The final step is critical. As Hirleman (1988) points out. many instruments will continue to "merrily report erroneous data and not notify the

user.'' An instrument operating envelope will be defined by the ability of the instrument to measure the desired property over an appropriate range to an acceptable accuracy. Hirleman (1988) groups the parameters which comprise the operating envelope into three domains: particle, instrument, and environmental properties. A general overview of the operating envelopes of in situ methods follows.

Particle Properties

A variety of particle properties can be of interest, including size, shape, concentration. velocity, and refractive index. Each of these properties can be distributed among a population of particles. and the problem becomes one of measuring the related dismbutions. A further complication arises as all particle properties can show spatial or temporal variation. Measurement of particle size distributions demands that both particle sizing and counting be accomplished with great accuracy. High spectral resolution is required when the size distribution is itself of fundamental importance. for example, in understanding or predicting physical processes or in identifying origin or formation mechanisms. Ideally, the selected instrument's sizing range should suitably span the actual particle size. range. This can complicate the characterization of wide dismbutions, as particle sizing over more than one order of magnitude in size is difficult to cover with one instrument in one configuration. The distribution's behavior at its tails can be important, particularly when transforming from a frequency to a mass weighted distribution. A second property of interest is panicle concentration: mass, area, and number per volume of gas are each of interest in some context. In most situations, it is impractical (or impossible) to characterize every particle present: thus, it becomes necessary to infer the true aerosol properties from a measurement of some subset. The particle velocity distribution can be important in understanding dispersal, transport, or flux. In some applications, the correlation between particle size and velocity is desired. Even when particle velocity is not of interest itself, it may be a limiting factor in system performance. For example. high speed particles can pose signal-processing and response-time difficulties in SPC's.

Instrument Properties

An accurate determination of a particle size distribution requires that the instrument must both size and count particles accurately. Hovenac (1987) describes factors which adversely effect SPC sizing and counting performance. Although both size and count sensitivity are crucial for ensemble techniques as well. the discussion is complicated by the averaging nature of the measurement. Perhaps the most difficult aspect of making an accurate in situ measurement is in defining the sample volume, as particle velocities and trajectories cannot be controlled as in a sampling-type instruments (Holve. 1980). This difficulty applies to both ensemble and SPC techniques, and can lead to both sizing and counting errors. For most in situ systems, the sample volume is determined by the intensity profile of the illuminating beam and by the geometry and characteristics of the receiving optics (apertures, stops, lenses, filters, etc.). Laser beam intensity nonuniformities within the sampling volume result in trajectory-dependent scattered intensity profiles for even monodisperse particles. For the common case of a laser beam with a Gaussian intensity profile. a particle passing through the axis of a laser beam will scatter more light than if it passed through the edge of the beam. Thus. a small particle passing through the beam axis and a large particle passing through the beam edge could give comparable scattering amplitudes ("trajectory ambiguity." Gouesbet and Grehan, 1988). For intensity-based SPC techniques, such multivalued response degrades instrument accuracy. Moreover. the combination of a nonuniform beam profile and photodetector sensitivity creates the situation where the effective sample volume becomes size dependent, e.g., small particles are detected only by passing through the central portion of the. beam whereas large particles are detected over a much larger cross section. Both ensemble and SPC in situ techniques can suffer this counting bias, and all SPC's require some form of sample volume correction (e.g., Holve and Self. 1959 Holve, 1980).

One of the key parameters of interest is particle size. Several issues arise with regard to particle sizing with in situ techniques: precision (repeatability). accuracy (resolution). sensitivity (lowest detectable size), and dynamic range. One requirement for suing precision is a monotonic response curve (intensity or phase versus size); unfortunately. light-scattering techniques are frequently multivalued due to Lorenz-Mie scattering effects. Variations in particle shape and refractive index effects can dramatically effect the shape of the response curve, and will limit system accuracy unless calibrations or calculations are performed with similar particles. Many in situ optical systems are

based on mar-forward scattering techniques. which minimize shape and refractive index effects. Trajectory ambiguity also degrades accuracy for intensity-based techniques. All optical *in situ* techniques require that the laser beam waist be 4-5 *times* the size. of the largest particle to ensure nominally uniform illumination over the particle's surface (Holve. 1980). Making the linear dimensions of the measurement volume much larger than the largest particles also reduces the fraction of particles that suffer edge *effects* (Holve and Self, 1979). Note that enlarging the measurement volume can increase coincidence *errors*, and so trade-offs must be made.

Lens imperfections, *misalignment. electronic* and photodetector nonlinearities, and other nonidealities can significantly degrade all aspects of system performance (Holve and Davis, 1985). Beam intensity fluctuations and system misalignment transients can impair both instrument precision and accuracy. As a rule of thumb, optical and signal processing limitations generally limit the dynamic size range that can be measured (with one instrument at one setting) to about a factor of 30. Instrument noise is frequently a limiting factor in determining dynamic range. and can also influence precision. accuracy. and sensitivity.

There is always a desire far improved instrument sensitivity. For *in situ* SPC's, a lower detection limit of about 0.3 μm is typical, although sampling type SPC's can currently detect panicles to about 0.05 μm. Knollenberg (1985) describes theoretical detection limits for SPC's. and shows that the limit is dominated by background scattering from stray light or gas molecules present in the sampling volume. High panicle concentrations can also limit system performance. For example. in SPC's this can lead to Coincidence. dead time, and intensity attenuation errors. Coincidence *occurs* when two particles occupy the measuring volume at the same time. which may be counted as a single large particle, resulting in both a sizing and counting error and consequently skewing the *size* distribution to larger sizes. Coincidence places an upper limit on the number concentration that can be measured without significant interference for a given system configuration. This upper limit has been *shown* to be proportional to the probability of *interference* and inversely proportional to the effective measurement volume (Holve. 1980). Dead time occurs when the electronics are not ready when an event occurs because a previous event is still being analyzed; dead time effects can reduce or skew the measured size distribution. High particle concentrations between the sample volume and the receiving optics can reduce the intensity of light scattered by the panicle. The resulting error in intensity-based techniques would be to undersize all particles. In ensemble systems, multiple panicle scattering occurs at high concentrations. In this *case*. measurements of the *size* distribution *become* concentration-dependent.

All of the techniques discussed in this review require sophisticated data analysis. and most require a full inversion or deconvolution to finally resolve the desired size and number distributions. Real-time ensemble instruments demonstrate the classic case of inverting a finite set of measured responses to infer an unknown distribution (Hirleman, 1988). For intensity-based *SPC* techniques, Holve *(1980)* has discussed the need to deconvolve the resulting intensity histograms to account for trajectory ambiguity and size-dependency of the measurement volume. Although *beam* intensity *variations* have minimal effect on particle Sizing with phase-Doppler techniques. corrections still need to be made to account for size-velocity correlations and size-dependent sample volumes when concentration is required. The importance of proper data analysis or inversion cannot be overemphasized.

Environmental Properties

Refractive index gradients along the optical path can cause. beam steering, with a resulting change in optical collection angles. The length of the optical path, and medium temperature and pressure gradients determine the extent of beam steering. Gas conditions (temperature, pressure. composition) also effect the gas refractive index. Laser systems are readily adaptable to high temperature environments, as they can mitigate the influence of high thermal radiation background. There are also practical *issues* like optical access and window contamination that must be considered. Also. application of optical techniques in environments with high ambient light levels can lead to *spurious measurements* unless suitably filtered.

SINGLE-PARTICLE COUNTERS INTENSITY-BASED

This first class of instruments sizes and counts individual particles as they pass through an illuminated sample volume. As the particles pass through this region. they scatter light which is collected over some solid angle by the receiving optics, and focused onto a photodetector. The particle size is determined by the peak intensity of the scattered light. A variety of such techniques are now available. and many reviews of the topic are available (Holve et al., 1981; Knollenberg. 1979. 1981; Hovenac. 1987). All of the limitations and concerns reported for SPC's in the overview apply to this class of techniques, including counting statistics at low concentrations and coincidence and dead time effects at high concentrations. In particular. nonuniformities in the illuminating beam can result in both sizing and counting errors for this class of equipment, and some form of correction (either hardware or analytic deconvolution) is required. Intensity-based techniques are particularly sensitive to environmental features which alter either illuminating beam or scattered light intensities, such as window contamination or high particle densities between the sample volume and collection optics.

Forward Scattering Spectrometer Probe (FSSP). (Particle Measuring Systems. Inc., (PMS), Boulder, CO) The FSSP models are aircraft mountable probes which size particles based on the intensity of forward scattered light as they pass through a laser illuminated sensing volume. The newer model FSSP-300 provides better sensitivity (down to 0.3 μm) and higher resolution (31 channels) over its range (0.3 to 20 μm) than the mechanically identical FSSP-100 (15 channels over several size ranges, such as 0.5-8.0 and 5.0-95 μm). The velocity operating range for the instrument is from about 10 to 125 m/s. In the standard configuration, a particle velocity distribution is not measured by the FSSP. A multimode He-Ne laser beam is used to help improve response monotonicity by diminishing the Lorenz-Me regime oscillations. The system has been used extensively in characterizing clouds and fogs (Knollenberg, 1981). The operating principles and limitations of the FSSP have been described extensively (e.g., Knollenberg. 1981). A patented dual-detector arrangement is used to size only those particles passing through a prescribed sampling volume. Briefly, the sampling volume between two probe tips is illuminated by a laser from one of the tips. When a particle enters the volume, it scatters light which is collected by optics located in the other probe tip. While a dump spot blocks the main beam, the forward scattered light enters a beam-splitting prism and is focused onto two photodetectors. The signal photodetector is unmasked and reports an intensity maximum used to size the particle, while the annulus detector is masked to eliminate light from in-focus. centered particles. A comparison between the two signals for each particle is used as an acceptance criteria: particles passing far from the focal plane scatter a larger proportion of light into the annular detector and are rejected. Baumgardner et al. (1990) recently published a thorough review of the optical and electronic limitations of the technique. and provided an extensive bibliography of related publications. Issues addressed include sample volume, sizing, and counting uncertainties. Both concentration and sizing uncertainties are found to be quite large (both about 27%).

Polytec Optical Aerosol Analyzers. (Polytec GmbH, Waldbronn, Federal Republic of Germany; and Polytec Optronics, Casta Mesa, CA) The HC series particle sizers detect white light scattered at 90° by single particles to measure the number distribution in the approximately 0.4 to 100 μm volume equivalent (geometric) diameter range (Umhauer. 1983). The choice of white-light illumination is intended to maximize monotonicity of the scattering intensity vs. diameter response curve, and to reduce (though not eliminate) index of refraction effects. Several models are available, differing in optical geometry and. hence. in nominal size and concentration ranges. Particle size ranges available range from 0.4-22 μm (Model HC-2015) to 1.5-100 μm (Model HC-2470). The larger measurement volume required for the latter size range makes the HC-2470 more susceptible to coincidence errors, but the system is less susceptible to edge errors (Borho, 1970). Numerical correction for coincidence is possible; maximum concentration is about 1 6 particles/cc for the HC-2015 and 10^3 for the HC-2470. Both models classify particles into 128 size channels. with a dynamic size range of 1:30. The velocity operating range is from 0.1 to 10 m/s (optimal to 20 m/s); particle velocities are not measured. The HC series is suited to filter efficiency testing. especially at high pressures or temperatures, and is also widely used in pharmaceutical spray sizing.

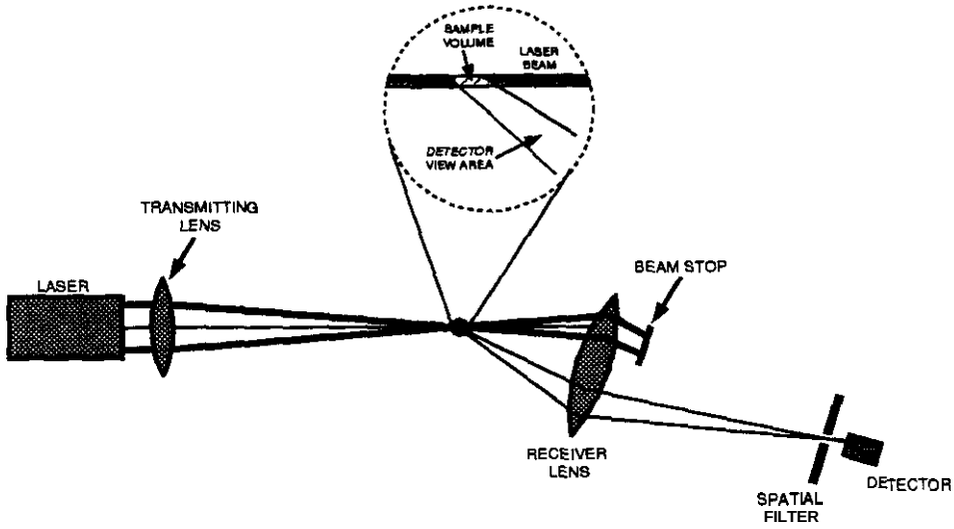

Figure 1. Layout of single panicle light scattering instrument (similar to PCSV).

Particle Counter Sizer Velocimeter (PCSV). (Insitec Measurement Systems, San Ramon, CA) The PCSV system is a single particle counter that measures particle size based on the intensity of He-Ne laser light scattered in the near forward direction (see Figure 1). Using near forward scattered (predominately diffracted) light helps reduce particle shape and refractive index effects: thus, instrument response is mainly dependent on particle cross-sectional area. The mean partide velocity is determined by averaging the widths of the scattered light pulses. The instrument's operating envelope is given by the manufacturer as: partide size between about 0.2 and 200 μm, concentration up to 10^7 particles/cm^3 for submicron and up to 100 ppm by volume for supermicron particles, and particle velocity between 0.1 and 400 m/s. A maximum particle pulse rate of 500 kHz is claimed for the system. To cover the wide range of sizes. two separate laser beams are used to form two independent measurement volumes: the narrower beam (nominal diameter of 20 μm) is used for sizing smaller particles, while the wider beam (nominal diameter of 200 μm) is used for sizing larger particles. Insitec claims an accuracy of +/-10% and a precision of +/-5% of the indicated size. An *in situ* alignment system is used to correct for beam steering in hostile environments (Holve and Annen, 1984). Alignment sensitivity was explored analytically by Holve and Davis (1985). A major feature of the PCSV system is the use of a deconvolution of the measured scattered intensity histogram to account for trajectory ambiguity and size-dependent measurement volume in determining the size distribution (Holve and Self. 1979 Holve and Annen, 1984; Holve and Davis. 1985). Lorenz-Mie scattering theory is used to predict the scattering response function (scattering intensity versus particle size) for the desired geometry, and has been experimentally confirmed (Holve and Self. 1979). The accuracy of the deconvolution algorithm was established using monodisperse droplets (Holve aad Self, 1979). Near real-time output is provided via a dedicated personal computer which performs the deconvolution. Insitec provides a rotating chrome-on-glass reference reticle for instrument calibration. PCSV system have been used to measure particle size distributions in a number of applications including coal/water slurry combustion (Holve and Annen, 1984). liquid fuel droplets and solid coal particles under combustion conditions (Holve. 1980), and soda-lime glass beads in both cold and hot flows (Holve and Self, 1979).

Uniform Beam Scattering: Laser Doppler velocimetry (LDV) is a well-established and documented technique for noninvasive measurement of particle velocities. made by measuring the Doppler-shifted frequency of light scattered by individual particles passing through a laser beam-defined measurement volume. The most common LDV configuration uses crossed laser beams to define a measurement volume with typical dimensions of the order 1 mm or less. Particles passing through the measurement volume scatter light with a Doppler shift proportional to the particle speed. Speeds as high as several hundred meters/sec can be measured using conventional electronics. The scattered light intensity signal from each particle passage ("Doppler burst") has the characteristic shape shown in Figure 2.

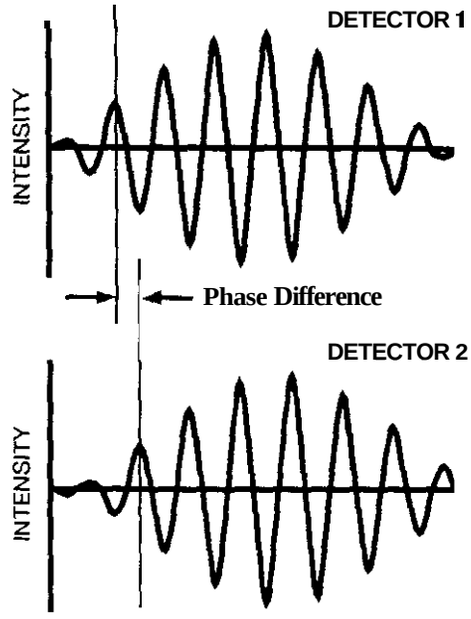

Figure 2. High-pass filtered laser Doppler velocimeter signals. The temporal frequency is directly related to the particle velocity

Ideally, an LDV system could be used for particle sizing, with the peak intensity of the Doppler burst directly related to particle size. However, the Gaussian nature of the laser beams limits the usefulness due to the ambiguity. In order to avoid use of deconvolution algorithms, so that individual particle size and velocity can be accurately, techniques have been developed which modify the incident beam intensity profile. This section describes and demonstrates measurements made with a dual system using a small pointing beam inside a larger laser beam of different wavelength. The pointing beam is used to find a relatively uniform portion of the large laser beam. Particles are only sized when detected by the pointing beam, ensuring that they are in the uniform portion of the larger beam, so the scattered pedestal intensity can recorded and the size calculated free of intensity ambiguity. The TSI 2000 and 200 (Meta Irvine, CA) instruments use a similar technique to provide a uniform measurement region for combined LDV, particle sizing, and velocity. The system can be configured to size through glass in cases where the particles are of unknown material or refractive index, and is designed to operate with no dead time, by digitizing of particle scattering events before processing the raw data. The manufacturer-stated operating envelope includes a 0.4 to 5000 μm size range, with 2% typical resolution and a 30:1 dynamic range, and a velocity range up to several thousand m/s. Data rates up to 3×10^6 particles/sec can be measured.

SINGLE-PARTICLE COUNTERS: PHASE

The phase Doppler technique is a laser Doppler velocimeter (LDV) based method for simultaneous measurement of size and velocity. This technique is not intensity-dependent like the previous group of SPC techniques. It can provide superior performance by minimizing effects such as beam attenuation or window fouling. A single point measures the spatial and temporal frequency of the Doppler-shifted light scattered by individual particles passing through a fringe pattern formed by beam-crossing measurement volume. Phase Doppler systems use multiple photodetectors to sample slightly different portions of the light scattered by individual particles. Figure 2 depicts typical high-pass filtered Doppler bursts measured by two spatially separated detectors. The phase difference between the two signals is a measure of the scattering angle spatial frequency, which is directly related to the particle refractive index and receiver configuration (Bachalo and Houser, 1984; Saffman et al., 1984). Particle velocity is related to the temporal frequency in the same manner as in conventional LDV. Figure 3 is a schematic layout of a generic phase Doppler system. Particle sphericity is required since the phase shift is calculated for either rays refracted

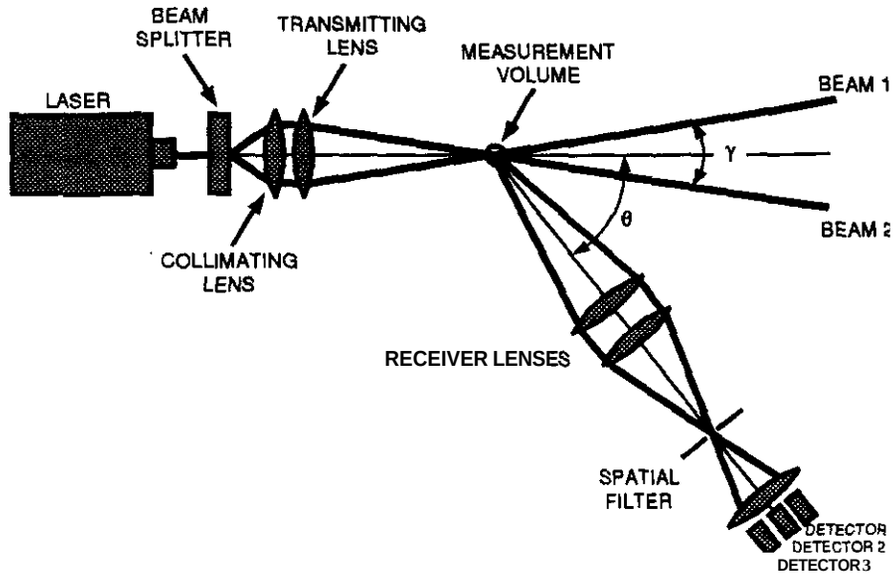

Figure 3. Layout of single particle phase Doppler instrument (similar to PDPA).

through spherical particles of known, constant refractive index or reflected off the surface of reflective particles. although preliminary work on measurement of nonspherical particles shows some promise (Alexander et al.. 1985).

Commercial instruments based on the phase Doppler technique include the phase Doppler Particle Analyzer (PDPA). (Aerometrics, Inc., Sunnyvale, CA) and the Particle Dynamics Analyzer (PDA, Dantec, Inc., Skovlunde, Denmark). The PDPA manufacturer-stated operating envelope includes a 1 to 8000 μm diameter range. with 5% typical accuracy and a 35:1 dynamic range, and a velocity range from 1 to 200 m/s, with 1% typical accuracy. This system also calculates number density based on the number of particles passing through a calculated size-dependent measurement volume (to correct for trajectory ambiguity effects). The maximum measurable number density is 10^6/cc. The PDA is described. with applications. in Saffman et al. (1984). This system includes a built-in laser diode is used to generate signals for calibration of the receiving optics. and signal phase is measured using a cross-correlation technique. The manufacturer-stated operating envelope for the PDA system include a 0.5 to 10,000 μm size range, with 4% typical accuracy and a 40:1 dynamic range. and a velocity range to greater than 500 m/s, with 1% typical accuracy. No number density measurement capability is currently claimed.

There has been an abundance of recent work with phase, Doppler instruments, both in instrument performance characterization and in applications in industrial and research settings. Jackson (1990) presents an overview of the PDPA instrument, Dodge et al. (1987) discuss liquid droplet measurements in sprays with the PDPA, and McDonell and Samuelson (1990) performed an evaluation of the sensitivity of PDPA measurements to operator input parameters. Dressler and Kraemer (1990) calibrated the PDPA using a multijet droplet generator. Ceman et al. (1993) performed a detailed investigation into PDPA droplet Sizing performance in the 3.5 to 25 μm range using a vibrating orifice aerosol generator and several different PDPA receiver geometries. They found nonlinearities in PDPA measurements of panicles below some critical diameter. in agreement with previous calculations (Al-Chalabi et al., 1988). Saffman et al. (1984) found that a large receiver lens collection solid angle tended to damp these oscillations. Recent work by Aerometrics (Sankar et al.. 1991) indicates that reflections from the surface of the droplet may also contribute to the oscillations in the phase versus diameter curve in the smaller diameter regime. Their work indicates that reflections can be minimized and linearity attained by collecting light at an angle close to the droplet Brewster angle. The results of Ceman et al. show that droplet sizing oscillations can be substantially reduced by using a larger collection solid angle (shorter focal length receiver lens), but that working near the droplet Brewster angle provided little or no additional improvement

SINGLE-PARTICLE COUNTERS IMAGING

Determining a particle's properties by direct imaging is among the earliest techniques used in particle measurement: consider the optical (and subsequently the electron) microscope. A significant advantage is that the shape and index of refraction issues which complicate single-particle scattering measurements are avoided. In fact imaging techniques provide one of the few avenues for investigating particle shape. The accuracy of single-particle imaging systems m limited by Fresnel diffraction and depth of field effects (Hovenac. 1987). Fresnel diffraction blurs image edges and complicates sizing. Depth of field effects arise from its dependence on particle size, with the result that large particles remain in focus over a greater axial distance than smaller ones.

Knollenberg (1979. 1981) designed an automated. *in situ.* single particle, optical imaging system which is commercially available (particle Measuring Systems. Inc., Boulder, CO) as the Optical Array Imaging Probe (OAP). In this family of probes, a collimated laser beam defines a measurement volume located between two sensing tips that extend forward from the main body of the system. Receiving optics direct the beam to illuminate a linear array of photodiodes. A particle passing through the measurement volume casts a shadow on the array, resulting in a decreased signal from the individual elements that lie in the shadow. In a I-D OAP system. the array elements are read and latched during the particle transit in a way that only provides particle size information. In a standard 2-D OAP system, the entire two-dimensional image of the particle is stored in high-speed memory as a series of "snap-shots" of the particle during its transit The great advantage of acquiring two-dimensional panicle images becomes apparent when measuring nonspherical particles. In the Grey Probe 2-D OAP system. a 64-element array is used where each element reports one of four shadow levels. The increased sophistication of the Grey Probe provides twice the resolution (twice as many elements) of the standard 2-D system. as well as providing depth-of-field information. For all of these imaging systems, both instrument resolution and sizing range depend on physical spacing of the array elements (typically 200 μm), magnification. and particle velocity. The latter requires that the user identify the expected velocity range in order to configure an OAP system. In addition. all of these systems reject particles which shadow elements at the edge of the array, as the fraction of the particle falling outside the array cannot be determined and thus precludes correct sizing. Depth-of-field rejection criteria can also be set for the grey probes. requiring that the particle must shadow at least one array element greater than a Specified level to be recorded. One difficulty is that high speed particles may not give a sufficiently dark shadow (due to electronic limitations) to meet this acceptance. criteria.

Particle sizing ranges and resolution depend on the particular model. which differ in the number of array elements and optical configuration used. The cloud-droplet models are suited to sizing smaller particles, with ranges such as 10-620 μm (with 10 μm resolution) or 200-6.000 μm (with 200 μm resolution). Precipitation models are suited to sizing larger particles. with ranges such as 50-3,100 μm (with 50 μm resolution) or 150-9300 μm (with 150 μm resolution). The resolution limits given above assume instrument operation at aircraft speeds (100 m/s); significant improvement in instrument resolution can be achieved at lower velocities. The lower limit for OAP sizing (somewhere between 1 and 10 μm according to Knollenberg. 1979) results from sampling considerations that result from the vanishingly small depth-of-field at these sizes. Particle velocities are not measured explicitly, but could be recovered by later analysis of the image sequences using the known imaging frequency. A ground-based precipitation OAP is available in either the 1-D (droplet sizing range 200-12,400 μm with 200 μm resolution from 62 size channels) or grey probe (droplet sizing ranges 200-12,400 or 70-4340 μm with 200 or 70 μm resolution. respectively. from 62 size channels) configuration. The distance between probe sensing tips is 50 cm which provides a large sampling ma.

ENSEMBLE TECHNIQUES PARTICLE FIELD IMAGING

Imaging systems are useful since they can "freeze" the motion of a particle-laden flow, allowing later analysis and providing a permanent record of transient events. In addition. imaging techniques make no assumption of sphericity. and can be used to allow visual examination of the shape of individual particles, along with providing statistical information on the particle size dishibuticm. concentration. velocity distribution. etc. Imaging measurements provide a primary measurement standard. since individual particle sizes are measured directly. In addition, an archival record

of the particle field is formed, allowing subsequent examination by various means. However, field imaging techniques do not provide real-time analysis and imaging resolution is often degraded by dense partide fields, window effects, and other common conditions. A number of imaging techniques are available for measurement of particle fields. including photography and holography (often combined with image processing).

Photographic investigations are attractive since the technique is well established and high resolution is possible. Photography of small particles places special restrictions on the process. often requiring very short exposure times to avoid blurring of moving particles and high resolution to detect the size range of interest. The achievable resolutions of photographic systems is often sufficient for particle measurements: however, the depth of field suffers. Image processing techniques promise. to improve the speed and accuracy of photographic studies, and have been applied in numerous studies (e.g., Oberdier, 1984). A commercial instrument using ensemble imaging is the Model 700 Particle and Spray Analyzer (Greenfield Instruments, Greenfield, MA). The manufacturer-stated operating envelope for this video-based instrument includes a 3 to 18500 µm diameter range. with a 150:1 dynamic range.. and the capability to measure nonspherical and opaque droplets. Velocity is not measured

Holography has become a fairly common technique for the study of particles with diameters larger than about 5 µm. A hologram is an interference pattern formed by the mixing of two coherent wave components; a subject wave reflected or scattered from the object or field of interest. and a reference wave. Holographic reconstruction creates three-dimensional images of the original illuminated volume. which can then be examined in detail for particle size and shape. as well as velocity and acceleration in systems with multiple-pulse illumination. The three-dimensional aspect allows simultaneous matching of the requirements for high resolution and good depth of field. unlike the photographic process. In addition, image pre-magnification (before recording) can allow examination of smaller particles, at the cost of reducing the sampled volume. Reviews of the use of holography for particle field measurements are given in Thompson (1974). Trolinger (1975). and Tyler and Thompson (1976). Holographic images of particle fields contain information on the size, shape and three-dimensional spatial position of each individual partide comprising the field.

After recording, the hologram is mounted in a reconstruction system, where it is illuminated with another coherent light source, acting as the conjugate of the original reference beam. The hologram acts as a diffraction grating to form three-dimensional real and virtual images of the original sample volume. Use of translating stages allows detailed examination of the reconstructed image. Holographic reconstruction and detailed data acquisition are very time-consuming. often requiring on the order of several "man-days" for examination of the several hundred particle images needed for statistical significance. Examinations are typically performed by visual observation of the reconstructed images. although automated analysis techniques are a topic of great interest (e.g., Haussmann and Lauterborn, 1980. Schafer and Umhauer. 1987, Chavez and Mayinger, 1990). Ewan et al. (1984) and Hess and Trolinger (1985) describe unique applications of the Malvern diffractometer (see below) for evaluation of reconstructed holographic images. Instead of measuring the size distribution of a particle field. the Malvem is set up to examine a particle field hologram, yielding ensemble size distributions for each region probed by the Malvern laser beam. Use of the Malvern device could be a very useful step toward automated reconstruction (at least in an ensemble sense). While no off-the-shelf holographic aerosol measuring systems are currently available (to the knowledge of the authors). several companies can design and install custom holographic systems. Included are MetroLaser (Irvine, CA) and Physical Research. Inc. (Torrance, CA).

ENSEMBLE TECHNIQUES FRAUNHOFER DIFFRACTION

Some of the first commercial. laser-based. in situ particle measurement techniques used Fraunhofer diffraction to characterize droplet sprays (Cornillault, 1972; Wertheimer and Wilcock. 1976 and Swithenbank et al., 1977). The technique determines a size distribution from a measurement of the ensemble diffraction pattern that results from the illumination of a particle cloud by a collimated laser beam. The technique has been developed into a variety of commercial systems which have been extensively characterized calibrated. and used in a wide range of particle studies. Excellent reviews of Fraunhofer diffraction techniques have recently appeared (Felton, 1990; Meyer and Chigier, 1986). In typical ensemble diffraction techniques Figure 4), a laser beam is expanded and then collimated

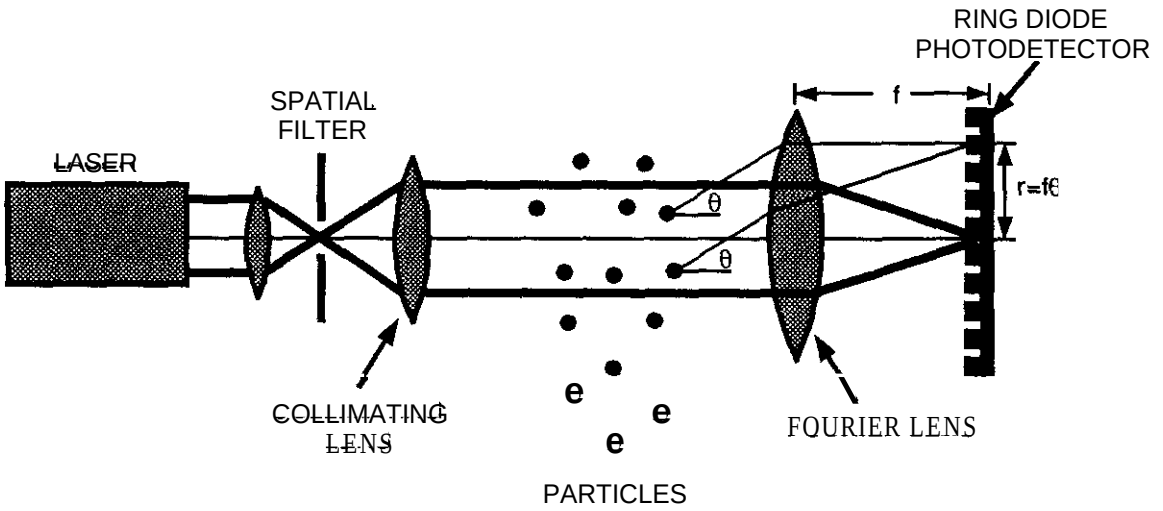

Figure 4. Layout of ensemble diffraction instrument.

into a beam several mm in diameter which passes through the particle cloud. Particles in the beam scatter light in all directions, although with particular efficiency in the near forward direction. A receiving lens is used to focus both the transmitted beam and forward scattered (predominately diffracted) light onto a detector located at the back focal plane of the lens. The transmitted light is focused to a point on the optical axis. while the diffracted light forms a series of concentric rings (Fraunhofer diffraction pattern). In general. smaller particles scatter light to a wider scattering angle 0. As the receiving lens performs a Fourier transform on the scattered light, light scattered at a given angle θ by a particle located anywhere in the illuminated sample volume will be focused at the same radial position in the transform plane. Thus, the resulting pattern is unaffected by particle location or motion. As a practical concern, the entire particle cloud should be within the focal length of the receiver lens. If particles are farther away than this. they may scatter light beyond the receiving lens: this effect is called "vignetting" (see Hirleman et al. 1984). Vignetting tends to bias the distribution towards larger sizes. as the small particle (large angle) signal is preferentially lost. The ensemble diffraction technique results in the classical inversion problem wherein a continuous size distribution is sought which provides best agreement with a finite set of experimental measurements (the scattered energies measured by each annular detector). A description of the inverse problem as it applies to ensemble diffraction techniques is given by Felton (1990) and Hirleman (1987). Typically, an iterative solution is used that minimizes the sum of squared errors between the predicted and measured detector responses. Once the size distribution is determined. the panicle concentration is determined by a direct measurement of the beam attenuation. No measurement of particle velocity is made by ensemble diffraction techniques.

Implicit in the standard analysis of ensemble diffraction techniques is the assumption that the particle diameter is much larger than the wavelength of the illuminating light. Thus. the lower limit for an ensemble diffraction instrument (for visible light) is about I μm. As an upper limit, diffraction from large particles (above several hundred microns) is concentrated at very small angles near the axis of the illuminating beam which makes measurement difficult. A second common assumption in the standard analysis is that each photon undergoes at most one scattering event while passing through the cloud. For high concentrations or long path lengths, multiple scattering spreads light over a wider angle with the result that the sue distribution appears wider and is skewed toward smaller sizes. Both experimental (Dodge, 1984a; Felton, 1990) and theoretical (Felton, 1990) studies of multiple scattering effects have been reported. Most studies agree that multiple scattering effects can be safely ignored for obscurations (fraction of the beam which is scattered by the panicle cloud) below about 50% (e.g., Felton. 1990; Meyer and Chigier, 1986). For higher obscurations correction factors and empirical correlations can account for multiple scattering (Felton. 1990; Dodge, 1984). Using analytic models or experimental correlations. it is possible to correct measurements at

obscurations as high as 98% (Felton, 1990).

Several other general limitations of ensemble diffraction techniques should be discussed. First. the technique implicitly assumes a uniform light intensity, while the hue radial intensity profile is Gaussian. As a result, the measurement volume for small particles will be smaller than for larger particles, thus biasing the measurement towards larger sizes (Meyer and Chigier, 1986). Second. beam steering can result when measurements are made through regions characterized by high (particularly time varying) gradients in index of refraction. The result is similar to system misalignment where the beam begins to illuminate the inner rings resulting in a false large particle response. Miles et al. (1990) offer an approach to making measurements in systems with time varying refractive index gradients. Finally, the ensemble technique gives a line-of-sight measurement with little or none spatial resolution. "Tomographic" techniques for deconvolving line-of-sight data to obtain radial profiles (for axially symmetric sprays) have been discussed by Hammond (1981) and Zhu et al. (1987).

Although of general importance to all ensemble diffraction techniques, most of the results summarized above were obtained with Malvern Instruments systems (Malvern Instruments, Inc., Southborough, MA. and Malvern Instruments Ltd., Worcestershire, UK). Other commercially available ensemble diffraction techniques are offered by Insitec (San Ramon, CA), Compagnie Industrielle des Lasers (CILAS, Marcoussis, France), Leeds and Northrup Instruments (Microtrac Division. FL), and The Munhall Company (Worthington, OH). Typical size ranges for these systems are 0.5 to 2000 µm, dependmg on configuration.

PERFORMANCE VERIFICATION

Although the *in situ* optical techniques reviewed provide a powerful resource for the measurement of particle size distributions. care must be taken to ensure that they are only used within their proper operating envelope. Thus, a means of verifying instrument performance - a standard - is essential to order to characterize instrument limits and to identify inherent instrument bias and inaccuracies. Moreover, Scarlett et al. (1990) emphasize that performance verification must include a standardization of operating procedures, as these can have a significant impact on instrument performance. Thus, a standard must include a document that prescribes a test method as well as a standard reference material (SRM) that is precisely characterized (Scarlett et al., 1990). Hemsley et al. (1988) and Rader and O'Hern (1993) discuss instrument calibration using SRM's (well-characterized beads or powders. reticles, or droplet generators), as well as instrument comparisons used to verify performance.

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