

The statistical convergence of aerosol deposition measurements

J. C. Moran, J. H. Lienhard V

375

Abstract The spatial distribution of deposited aerosol particles shows substantial statistical variation. This variation is especially important for sampling techniques having a high spatial resolution, such as laser-based methods, or for experiments involving a small sample size (few identical locations measured) or a short duration. This paper develops a criterion to ensure adequate statistical convergence in deposition measurements. The time evolution of statistical variations is modeled using standard Poisson statistics; this model provides a bound on measurement uncertainties. The model is compared to experimental measurements of the spatial variation of deposition. Numerical simulations are used to examine the effects of aerosol polydispersity on statistical convergence. Both the experiments and the numerics show excellent agreement with the model.

List of symbols

Roman letters

A	area of one measurement
\bar{n}	sample mean number of particles deposited
N	number of measurements in a sample
n	number particles deposited in area A during time T
s_n	sample standard deviation of number of particles deposited
s_v	sample standard deviation of voltage from LIF deposition measurements
T	time duration of deposition process
\bar{v}	sample mean voltage from LIF measurements

Greek letters

λ	particle deposition rate per unit area
μ_n	true mean number of particles deposited in an area A during a time T
σ_n	true standard deviation of number of particles deposited in an area A during a time T

1 Introduction

Particle deposition measurements are made in broadly differing contexts. Particle deposition is one of the major problems facing the semiconductor industry, owing to a significant loss of product yield that results from microcontamination during manufacturing (Cooper 1986). Depositing particles may cause surface erosion in machinery (Smeltzer et al. 1969). Deposition in the lung of inhaled particulates has been studied extensively for its health effects (e.g. Chan and Lippman 1980; Cohen et al. 1990; Ferin et al. 1990) and has more recently been considered for drug delivery (Sweeney et al. 1990). Deposition measurements are frequently used to examine fluid dynamical models of deposition mechanisms (e.g. Abuzeid et al. 1991). Typically, a deposition measurement determines the number or mass of particles deposited per unit area of surface during a set interval of time.

Deposition of particles from a flow occurs randomly at each location along a surface bounding the flow. Consequently, the number of particles found at any location after a given length of time is a random variable, so that any single measurement at that time and position possesses considerable statistical uncertainty. The uncertainty is more severe for high spatial resolution measurements, where individual samples may include relatively few particles, and for experiments with low deposition rates or short run times.

Few papers on aerosol deposition address these spatial variations explicitly, and some traditional sampling techniques (such as dye washing) require relatively large surface areas for single measurements, so that spatial variations are immediately averaged out. Our interest in this question is driven by our efforts to apply a high-spatial resolution laser sampling technique (Nowicki 1994; Roth 1995) in which 0.5 mm^2 areas are measured and for which spatial variations have a strong impact on the quality of the data. For high resolution sampling techniques, it is vital to have an estimate of the statistical uncertainty in deposition data for given probe areas, deposition rates, and run times. In this paper, we apply standard statistics to obtain such an estimate.

We first model the time evolution of the mean and standard deviation of the number of particles deposited within a given area A for a given mean particle arrival rate λ . The statistical distribution may be viewed as that obtained by repeating the experiment N times for a single location or as that obtained by taking N samples at many locations in a spatially homogeneous flow. Experimentally, A represents the area over which a single deposition measurement is made (0.5 mm^2 for the laser probe

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J. C. Moran, J. H. Lienhard V
Warren M. Rohsenow Laboratory
Department of Mechanical Engineering
Room 3-162
Massachusetts Institute of Technology
Cambridge, MA 02139-4307, USA

Correspondence to: J. H. Lienhard V

used herein). The mean deposition rate, λ , is set by characteristics of the flow field from which the particles deposit, and it usually varies significantly with position along the surface. For our purposes, it is sufficient to treat λ as having negligible variations over the small area A (so that gradients of the flow occur on a scale large compared to A).

To test this model, we have performed measurements of the spatial distribution of the number of particles deposited from a homogeneous turbulent channel flow onto a flat wall. The test aerosols are liquids doped with a fluorescent dye. Spatially localized samples of the deposited liquid are obtained using a fiber-optic probe to excite and receive fluorescent light from the dyed liquids. The distributions so obtained are analyzed statistically. Test aerosols were either water containing fluorescein dye or a lubricating oil containing commercial test dye.

Numerical simulations of the deposition process were also performed. Particles were randomly deposited into the cells of a test matrix, with each cell in the matrix representing a measurement location. After a given number of particles were placed in the matrix, the spatial distribution and its statistics were obtained by analyzing the contents of the matrix. The numerical model was used to examine the impact of aerosol polydispersity on the rate of convergence of the statistics.

2

Statistical model

The statistical model for the time evolution of aerosol deposition is based on the Poisson distribution, which will apply if the conditions of the experiment are constant in time and the number density is low enough that successive deposition events are independent. Under the Poisson distribution, the probability of n particles being deposited at a specific location on a surface during a time interval T is

$$p(n, \lambda TA) = \frac{(\lambda TA)^n \cdot e^{-\lambda TA}}{n!} \quad \text{for } n=0, 1, 2, 3, \dots \quad (1)$$

The mean of the distribution, or true mean, is the average value of n that would be obtained from an infinite number of measurements having identical A , λ and T ; its value is $\mu_n = \lambda AT$. In practice, a finite number of measurements, N , is taken and the sample mean, \bar{n} , is computed. The sample mean, \bar{n} , differs from the true mean, μ_n , owing to statistical variations in the deposition process, which have a variance $\sigma_n^2 = \lambda AT$. We may gauge the size of this variation by considering the ratio

$$\sigma_n / \mu_n = 1 / \sqrt{\lambda AT} \quad (2)$$

The role of the standard deviation to mean ratio in bounding the experimental error can be found by using a confidence interval to estimate the percentage error in the mean deposition of a set of measurements. A 95% confidence interval for the true mean μ_n is obtained from the normal distribution (Beckwith et al. 1993):

$$\bar{n} - \frac{1.96 \sigma_n}{\sqrt{N}} \leq \mu_n \leq \bar{n} + \frac{1.96 \sigma_n}{\sqrt{N}} \quad (3)$$

Here, N is the number of measurements used in computing the sample mean \bar{n} and the sample standard deviation s_n . It follows

that the fractional error in the sample mean is bounded as shown

$$\frac{|\mu_n - \bar{n}|}{\mu_n} \leq \frac{1.96 \sigma_n}{\sqrt{N} \mu_n} = \frac{1.96}{\sqrt{N}} \frac{1}{\sqrt{\lambda AT}} \quad (4)$$

at a confidence level of 95%. If only one measurement is made, a fair *estimate* of the fractional uncertainty in that measurement (at a 95% level) is $(2\sigma_n / \mu_n) = 2 / \sqrt{\lambda AT}$.

Equation (4) provides a means of estimating the values of experimental parameters required to limit statistical uncertainties to a desired level. In a typical case, the area A will be set by the specific experimental method, and a ballpark value of λ may be known. If the time duration T of the experiment is constrained by other factors, then Eq. (4) can be used to estimate the required sample size N . If, on the other hand, the number of identical measurements N is limited, Eq. (4) prescribes the run time needed to achieve a desired statistical uncertainty. As a specific example, suppose that a particular experiment has $\lambda A = 0.0015$ particles/s, and that an uncertainty in \bar{n} of 10% is desired. If 210 samples are to be taken, Eq. (4) shows that a run time of 20 min is required. If, instead, only *one* measurement were to be taken ($N=1$), a run time of over 70 h would be required.

3

Numerical simulations and polydispersity

Numerical simulations of the deposition process were run in order to examine the influence of aerosol polydispersity. Polydispersity in a particular concern because the distribution of particle size introduces a second source of statistical fluctuation that is not accounted for in the model of Sect. 2.

A random number generator was used to assign particles to random cell within a 32913 element matrix at an effective rate of 50 particles/s, so that $\lambda A = 50/32913 = 0.0015$ particles/s. As would be expected, the resulting distribution of particles among the cells of the matrix showed excellent agreement with a Poisson distribution. At intervals of 20 s, the number of particles in each of 210 cells was recorded and the mean and standard deviation of this 210 point sample was calculated.

To incorporate polydispersity, the particle size distribution of Fig. 1 was used; this distribution corresponds to our experiments in Sect. 4 below. The depositing particles were scaled into diameter bins in proportion to the distribution of Fig. 1 and deposited randomly into the matrix. The deposition for each cell was computed as the sum of the diameters of all particles deposited in the cell, and a sample mean deposition was calculated by averaging the deposition in a sample of such cells. The sample standard deviation of deposition was computed in a similar way. The ratio of sample standard deviation to sample mean is shown in Fig. 2 as a function of time for a sample of $N=210$ cells. Figure 3 shows the evolution of this ratio for a sample of just $N=9$ cells.

In these figures, the scatter of the computed data represents the statistical fluctuations that result from the randomness of deposition; each plotted point is the possible outcome of an experiment having the given sample size. The polydisperse data differ little from the monodisperse data, suggesting that the additional variability introduced by particle size variations is small. These figures also illustrate the effect of run time and

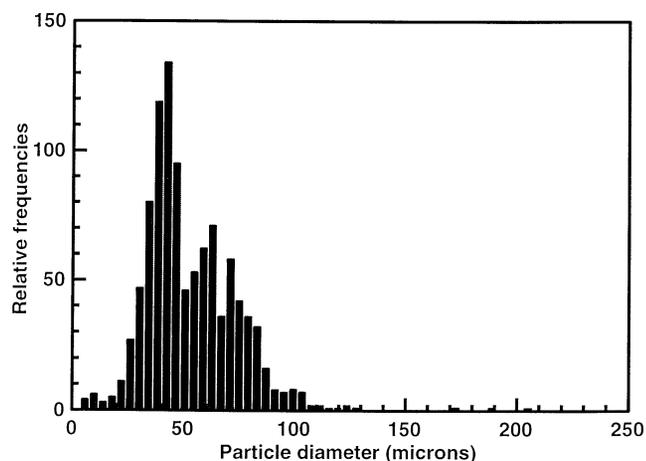


Fig. 1. Aqueous aerosol size distribution produced by the array generator

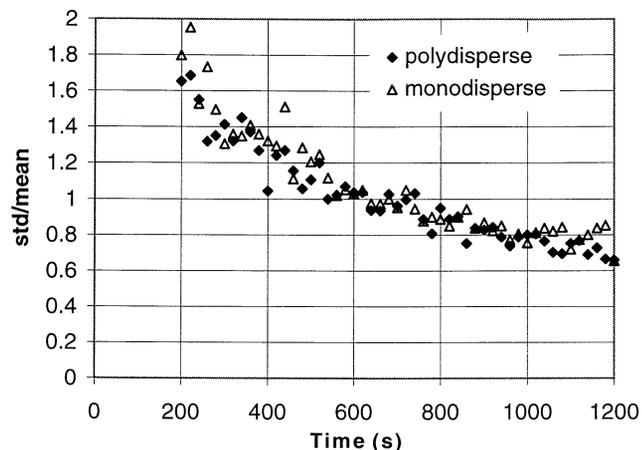


Fig. 2. Numerically generated ratio of sample standard deviation to sample mean for a sample size of $N=210$

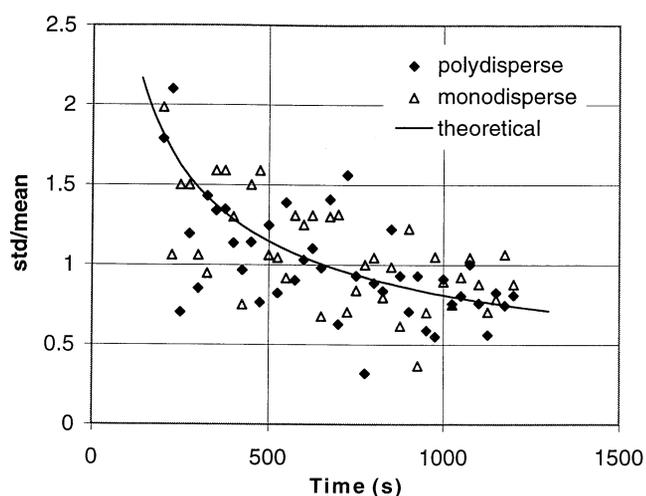


Fig. 3. Numerically generated ratio of sample standard deviation to sample mean for $N=9$. Theoretical curve calculated from Eq. (2)

sample size on the statistical convergence. After 1000 s, the true standard deviation to mean ratio has a value of about 0.8 as calculated from Eq. (2) with $\lambda A = 0.0015$ particles/s. For a sample of $N=210$, (Fig. 2), Eq. (4) shows that the corresponding uncertainty of \bar{n} is just 11%. In contrast, for $N=9$ (Fig. 3), the uncertainty is 53%.

4 Experimental observations of statistical convergence

Deposition experiments were conducted in wind tunnel. An aerosol generator seeds the air flow in a settling section upstream of a 20:1 area ratio contraction which is followed by a 1.5 m long test section (Roth 1995). In the present experiments, the test section consisted of a long rectangular channel (5 cm by 25 cm cross-section) through which the aerosol laden flow passed at 10 m/s with a Reynolds number based on hydraulic diameter of 16000. Under these conditions, the flow in the channel is turbulent with an essentially homogeneous core outside the boundary layer. The boundary layer characteristics are invariant along the length of the channel (beyond a short entry region). The bottom surface of the channel was a flat black plastic upon which aerosols deposited.

A Laser Induced Fluorescence system (LIF) was developed in order to measure deposition over spatially localized regions (Nowicki 1994). The LIF system operates by illuminating a small mass of deposited aerosol on the wall of the test section with an argon-ion laser. The aerosol contains a known concentration of a fluorescent dye which absorbs the laser light and re-emits light at different wavelength. The intensity of this emitted light is proportional to the mass of aerosol excited by the laser; this enables the amount of aerosol in the probe area to be determined.

The exciting laser beam and the emitted light were carried by a fibre optic cable to a probe focused on an area $A = 0.5 \text{ mm}^2$ of the surface where deposition occurred. The emitted light was detected by a photomultiplier tube circuit whose output is recorded as a voltage by a computer. By moving the probe, many independent but identical points on the deposition surface could be measured to produce the sample size N desired. For the present experiments, a fixed set of $N=210$ locations were sampled within a 12 cm by 16 cm region of the bottom surface of the test section.

The LIF voltage measurements were used to compute a sample mean voltage \bar{v} and a sample standard deviation of voltage, s_v . These values are related to \bar{n} and s_n through multiplication by a calibration constant. For present purposes, however, it is sufficient to compute

$$s_n/\bar{n} = s_v/\bar{v} \quad (7)$$

4.1 Experiments with aqueous aerosols

The aerosol generator used for water aerosols was a two dimensional high-power acoustic droplet generator, or Array Generator (Dressler 1993). It consists of a vibrating transducer which drives the breakup into droplets of jets leaving an array of 225 orifices. The generator was placed in the settling section of the wind tunnel, 20 cm from the contraction. A Phase Doppler Particle Analyzer (PDPA, Aerometrics Inc.) was used to measure the aerosol's velocity and diameter distributions

within the test section. A typical diameter distribution is shown in Fig. 1. The particle volumetric mean diameter for this distribution was $58 \mu\text{m}$.

The water aerosols were doped with fluorescein at a concentration of 0.001% by weight and the illuminating laser power (at 488 nm) was kept constant at 0.5 mW. Aerosol number densities were kept at relatively high levels inside the channel to humidify the air and suppress evaporation of the aerosols. Water evaporation increases the concentration of fluorescein in a droplet, since the fluorescein does not evaporate, and thus results in a larger LIF signal. In this sense, the system has a natural evaporation compensating effect. However, to check for an influence of evaporation on the statistical convergence, experiments were also run using low-vapor pressure oil aerosols (see Sect. 4.2).

Experiments were performed for run times T ranging from 100 to 360 s. Experiments longer than 360 s were difficult to run owing to very high levels of aerosol deposition in the channel which led to rivulets on the surface as well as saturation of the PMT amplifiers. Experiments shorter than 100 s were difficult to run owing to a lack of sufficient aerosol deposition.

A typical aerosol deposition distribution is shown in Fig. 4 in terms of the LIF sample voltage. The sample mean LIF voltage for this experiment is 1.2 V and the sample standard deviation is 1.66 V. Treating these as good approximations to the true mean and standard deviation, Eq. (2) can be used to calculate λ , which for these experiments has an average value of 0.0023 particles/mm² s. With λ , the theoretical ratio, σ_n/μ_n , may be plotted as a function of T . This curve is shown together with the experimental data points, s_n/\bar{n} , in Fig. 5.

Error limits for the individual sample mean readings were calculated using a student t -test with a 95% uncertainty level. Error limits for the sample standard deviation were computed with the chi-square test. These were combined to produce the error bars displayed in Fig. 5. The theory shows close agreement with the data.

Figure 5 shows that the rate of convergence is very rapid for the first 50 s, but it then slows considerably. The standard

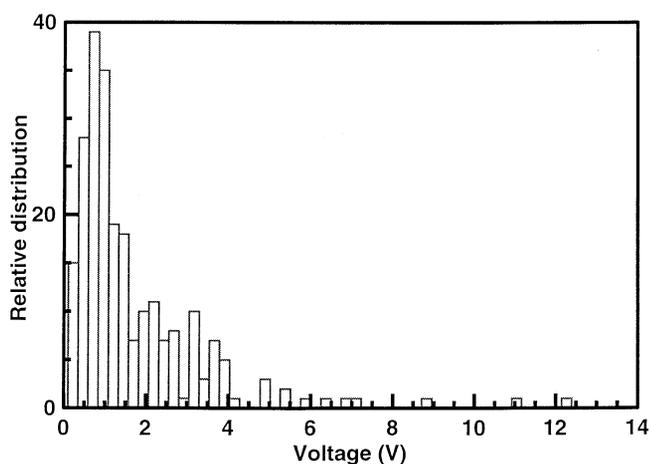


Fig. 4. Distribution of 210 voltages sampled by the LIF apparatus for aqueous aerosol deposition

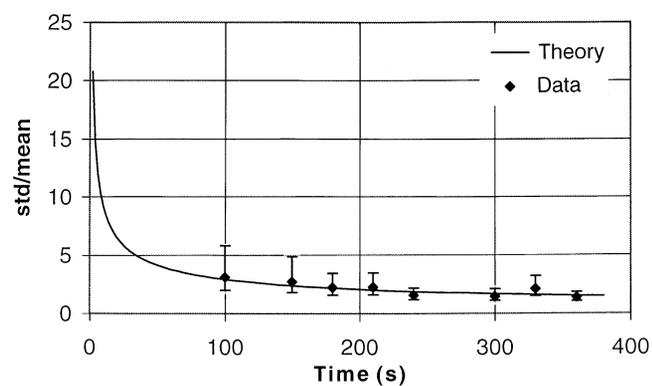


Fig. 5. Sample standard deviation to sample mean ratio measured for aqueous aerosol deposition as a function of run time. Theoretical curve calculated from Eq. (2)

deviation is still 1.5 times the mean after 6 min. In order for the standard deviation to be just 10% of the mean, an experimental run time of 24 h would be required. For a sample of $N=210$ points, Eq. (4) shows that a 10% uncertainty in the sample mean is reached after a run time of $T=26.5$ min. If, on the other hand, just *one* point was measured to obtain the sample mean, a run time of over 93 h would be required to get a 10% uncertainty. If the one point was measured after a 26.5 min run time, its uncertainty would be 145%.

4.2 Oil aerosol experiments

Experiments were also performed using a lubricating oil as the aerosol medium. This oil has a low vapor pressure (it is not susceptible to evaporation) and also has different surface wetting characteristics than deposited water aerosols. A commercial dye (Oil-Glo, Spectronics Corp.) was used to provide fluorescence. A compressed air nebulizer generated the aerosols (Yoon 1991). It operates by passing a high velocity air jet over a 6.3 mm diameter liquid supply tube, creating a low pressure region over the tube that draws liquid out of the tube and disperses it as a droplet cloud. The volumetric mean diameter of the oil droplets from the nebulizer is $100 \mu\text{m}$.

Experiments ranged in duration from 15 to 120 s. Experiments longer than 120 s were difficult to perform owing to film formation on the channel walls. The constant λ had an average value of 0.1172 particles/mm² s. With this value the theoretical convergence rate can be plotted using Eq. (2). The equivalent constant required for the numerical simulation is $\lambda A = 0.0683$ particles/s. The experimental, numerical and theoretical results are compared in Fig. 6.

All three results are in good agreement. The convergence rate is much faster than in Fig. 5, owing to the very large number of particles depositing per second. At these high deposition rates, the uncertainty in \bar{n} for a sample size of $N=210$ is 4.8% after a run time of $T=120$ s. If, on the other hand, just one probe area were measured ($N=1$), the uncertainty at this time would be 70%.

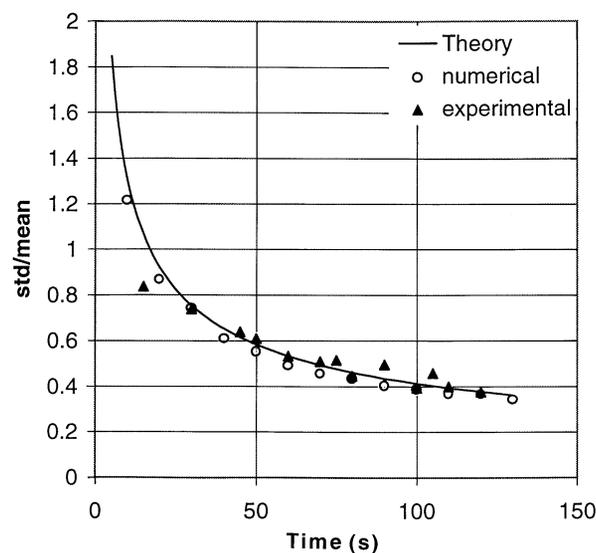


Fig. 6. Sample standard deviation to sample mean ratio measured for oil deposition as a function of run time

5

Conclusions

Statistical variations in the deposition of aerosols may lead to large uncertainties in measurements of aerosol deposition rates, particularly for measurements of high spatial resolution. These errors can be reduced by extending run times or by increasing the number of locations sampled. This study has developed a quantitative criterion for the statistical convergence of such deposition measurements.

1. If the number of particles deposited at a location is n , the ratio of true standard deviation, σ_n , to true mean, μ_n is given by Eq. (2). The percentage uncertainty in the mean number of particles, \bar{n} , in the measurement area A after a time T is

$$\left(\frac{1.96}{\sqrt{N}} \frac{1}{\sqrt{\lambda AT}} \right) \times 100$$

at a 95% confidence level. Here N is number of samples and λ is the rate of deposition per unit area.

2. Numerical simulations suggest that aerosol polydispersity has little effect on the statistical convergence rate.
3. The high-spatial-resolution LIF data for both water aerosols and oil aerosols are in good agreement with the theoretical results. The LIF probe area is only 0.5 mm^2 , and consequently, spatial variations in the data are quite pronounced. This stands in contrast to more traditional low-resolution methods.

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