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VARIABILITY OF TOTAL MASS AND OTHER MEASURES OF SMALL SAMPLES OF PARTICLES

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Abstract—Aerosol measurement frequently requires that an aerosol sample be withdrawn from its environment. The sampled mass is not strictly proportional to the sampled volume because mass is present in discrete entities. This introduces a fundamental variability in the estimated particle mass concentration, particularly important for small particle samples. The total amount of any particle measure, w, including mass, of the sampled particles is described by its coefficient of variation, CV. The results also apply for sampling a volume of liquid in which particles are dispersed, and can under some conditions be used for, e.g. analysis of weight per cent of asbestos in bulk powder, and microscopical analysis of total particle mass or projected area in a specimen. The fundamental coefficient of variation CV is also given for the special case of individual particle diameters having a log-normal distribution. Exact knowledge of the size distribution at the upper tail is critical in determining CV, as illustrated by calculating CV for a range of right truncated log-normal distributions. A mass variability equivalent diameter, MVED, is defined, by which the mass variability of a polydisperse aerosol can be described in terms of number variability (Poisson) of a monodisperse aerosol with diameter MVED. A population of airborne particles, sized by microscopy is used to show that in order to obtain CV < 10% for particle mass, a sample of this particular aerosol must contain an expected mass of 0.01 mg, and an expected number of particles, $N > 6500$, while only 100 particles would be needed if the measure was particle number. The variability is termed fundamental because it is the lowest achievable variability for given sample size and size distribution. This must be recognized, when determining overall uncertainty budgets for analytical procedures, including use of direct reading particle mass monitors for which a simple equation is given for calculating CV.

NOTATION

CMD	count geometric mean diameter
GSD	geometric standard deviation
CV_{ws}, CV_{ms}	coefficient of variation of total sampled particle measure, and particle mass, respectively
CV_v,	coefficient of variation of sampled volume
D	volume or mass equivalent particle diameter
MVED	mass variability equivalent diameter
MMD, MMAD	geometric mean of mass weighted diameter, and of aerodynamic diameter, respectively
N	number of particles in sample
N_0	number of particles in any unit volume
V	volume of sampled matrix
W, M	individual particle measure, and particle mass
W_s, M_s	total measure, and total particle mass of particles in the sample

Greek letters

β	ln (GSD)
λ	expected number of particles in a unit volume of the matrix
μ	ln (CMD)
κ	dynamic shape factor
ρ	particle density

INTRODUCTION

Aerosol measurement frequently requires that an aerosol sample be withdrawn from its environment. If the withdrawn volume of air is known, and the total amount of aerosol

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in the withdrawn sample is analyzed, the aerosol concentration in the environment or the concentration of an analyzed species can be calculated. Since the air is a continuum, the sampled air volume can, in principle, be determined with any accuracy. However, the sampled mass is not strictly proportional to the sampled volume because mass is present in discrete entities. This introduces a variability in the estimated particle mass concentration, an effect which can be particularly important for small particle samples.

Mass is only one particle measure. Total surface is another. To generalize, the total amount of any measure, w, of the particles in the matrix sampled will be characterized. W is a random variable, and will be described by its expectation value, E, and variance, Var, as well as the resulting coefficient of variation, CV_{ws}.

The results apply for many other situations. One is sampling a volume of liquid in which particles are dispersed. Another is analysis of, e.g. weight per cent of asbestos in a bulk powder, provided the powder in which the asbestos is distributed, can be treated as a continuum. A third example is microscopical analysis of particles of a given type in a specimen. The parent population is then all particles of this type in the specimen. The sample is the area delineated by the chosen number of fields.

SAMPLING

The matrix from which the sample is drawn, and the sampling procedure for which the model is derived has to be specified in general terms. The following assumptions are made:

(1) The matrix will be treated as a continuum.

(2) The total volume of the particles in any unit volume of matrix is only a very small fraction of the unit volume.

(3) A particle is said to belong to a part of a matrix if its center of gravity is within that part.

(4) The distribution of particles in the matrix is uniform. The distribution of number, N_0, of particles in any unit amount of matrix is then approximately a Poisson distribution, $P(x)$. The expected value of number of particles in a unit amount will be denoted λ.

(5) A certain amount of the matrix is sampled. The amount is measured as matrix volume, matrix mass, or matrix area.

(6) Sampling of an amount of matrix is random, i.e. any part of the matrix is sampled with equal probability.

(7) All particles in the sampled part of the matrix belong to the sample. That is, there is no change in their number during the sampling process.

In the following, a unit volume of matrix is considered, unless indicated otherwise. It is however straightforward to generalize to any measure of a matrix. From the assumptions it follows that the number of particles, N_0, in any unit volume is

$$N_0 \in P(\lambda). \tag{1}$$

For a given volume, v, of matrix sampled, the number, N_v, of particles in the sample has a Poisson distribution with expectation value $E(N_v) = v\lambda$, due to assumptions 2 and 4, i.e.

$$(N \mid V = v) = N_v \in P(v\lambda). \tag{2}$$

Notice that upper-case letters will be used to denote the stochastic variable, lower case for its realization.

COEFFICIENT OF VARIATION OF TOTAL PARTICLE MEASURE

Let w denote a measure defined for an individual particle, and let there be N particles in a sample consisting of the random volume V of the matrix. The total w-measure, W_s, of particles in the volume V is

$$W_s = \sum_{i=1}^{N} W_i, \tag{3}$$

where W_i denotes the w-measure of particle No. i in the sample ($i = 1, 2, \cdots, N$). V, N, and W_s in equations (2) and (3), are stochastic variables. Due to the assumptions (2) and (4), V and N are statistically independent, and the W_i are independent, identically distributed random variables, i.e. $E(W_i) = E(W)$ ($i = 1, 2, \cdots, N$). The following relations can be used for calculating expectation and variance:

$$E(X) = E(E(X|Y)), \quad \mathrm{Var}(X) = \mathrm{Var}(E(X|Y)) + E(\mathrm{Var}(X|Y)). \tag{4}$$

Equation (4) is derived from a more general expression (see e.g. Wilks, 1962). It does not assume independence between X and Y. In order to calculate the expectation $E(W_s)$, it is first noted that for given volume V

$$(W_s|V = v) = W_{vs} = \sum_{i=1}^{N_v} W_i, \tag{5}$$

where N_v is a random variable with distribution given by equation (2). The expectation $E(W_{vs})$ is thus

$$E(W_{vs}) = E(E(W_{vs}|N_v)) = E(N_v E(W)) = v\lambda E(W). \tag{6}$$

Using equations (4) and (6) one obtains for $E(W_s)$

$$E(W_s) = E(E(W_s|V)) = E(V\lambda E(W)) = \lambda E(V)\ E(W). \tag{7}$$

The variance of W_{vs} is calculated using equations (2) and (4)

$$\begin{aligned} \mathrm{Var}(W_{vs}) &= \mathrm{Var}(E(W_{vs}|N_v)) + E(\mathrm{Var}(W_{vs}|N_v)) \\ &= \mathrm{Var}(N_v E(W)) + E(N_v\, \mathrm{Var}(W)) \\ &= v\lambda(E(W))^2 + v\lambda\, \mathrm{Var}(W) = v\lambda E(W^2). \end{aligned} \tag{8}$$

Equations (4) and (8) give

$$\begin{aligned} \mathrm{Var}(W_s) &= \mathrm{Var}(E(W_s|V)) + E(\mathrm{Var}(W_s|V)) \\ &= \mathrm{Var}(V\lambda E(W)) + E(V\lambda E(W^2)) \\ &= \lambda^2 (E(W))^2 \mathrm{Var}(V) + \lambda E(W^2)\ \mathrm{E}(V). \end{aligned} \tag{9}$$

The coefficient of variation of total sample measure, CV_{ws}, is then

$$\mathrm{CV}_{ws}^2 = \frac{\mathrm{Var}(W_s)}{E(W_s)^2} = \frac{\mathrm{Var}(V)}{E(V)^2} + \frac{E(W^2)}{\lambda E(V)\ E(W)^2} = \mathrm{CV}_v^2 + \frac{1}{\lambda E(V)} \frac{E(W^2)}{E(W)^2}. \tag{10}$$

CV_v is the coefficient of variation of the sampled volume of the matrix and can, in principle, be made arbitrarily small, since the matrix is assumed to be a continuum. The coefficient of variation is for $\mathrm{CV}_v = 0$

$$\mathrm{CV}_{ws}^2 = \frac{1}{\lambda E(V)} \frac{E(W^2)}{E(W)^2}. \tag{11}$$

Equation (11) represents the minimum obtainable coefficient of variation for given sample volume and size distribution, and is thus termed the fundamental coefficient of variation. It can only be decreased by increasing the sample size. The parameter $E(W^2)/E(W)^2$ is given by the size distribution of the particle population in the matrix. When $\mathrm{CV}_v = 0$, $E(V) = V$; but the notation $E(V)$ will be used throughout to indicate, that the equations can be used in case $\mathrm{CV}_v > 0$, by adding the volume variability according to equation (10).

In case the w-measure is the mass of the individual particle, then $W = M$, and the expected total mass, $E(M_s)$, of the particles in the sample is

$$E(M_s) = \lambda E(V)\ E(M) \tag{12}$$

from which λ is obtained as

$$\lambda = \frac{E(M_s)}{E(V)\ E(M)}. \tag{13}$$

The expected amount of sample in equation (11) is given in terms of particle number. The corresponding equation, if the expected amount of sample is given in terms of mass, can be found by substituting equation (12) in equation (11). This gives

$$CV_{ms}^2 = \frac{1}{E(M_s)} \frac{E(M^2)}{E(M)}. \tag{14}$$

PARTICLES WITH LOG-NORMAL DIAMETER DISTRIBUTION

In this section, the fundamental coefficient of variation will be calculated for the case where the particle population has a log-normal size distribution. The diameter probability density function $f(d)$ is then

$$f(d) = \frac{1}{\sqrt{2\pi}\beta} \frac{1}{d} \exp\left(-\frac{(\ln(d) - \mu)^2}{2\beta^2} \right) \tag{15}$$

or in short

$$D \in \mathrm{LN}(\mu, \beta^2) \text{ or } \ln(D) \in N(\mu, \beta^2), \tag{16}$$

where

$\mu = \ln$ (CMD); CMD = count geometric mean,

$\beta = \ln$ (GSD); GSD = geometric standard deviation.

D is the mass equivalent diameter, defined as

$$D^3 = \frac{6M}{\rho\pi}, \tag{17}$$

where ρ is particle density.

In order to apply equations (11) and (14) for the log-normal case, some relations for the log-normal distribution will be summarized (see e.g. Aitchison and Brown, 1969) :

The expectation value $E(D)$ of D is obtained as

$$E(D) = \exp(\mu + \tfrac{1}{2}\beta^2). \tag{18}$$

The distribution of individual particle mass, $M = (\pi/6)\rho D^3$ is also log-normal with parameters

$$M \in \mathrm{LN}(\mu_m, \beta_m^2), \quad \text{where } \mu_m = 3\mu + \ln\left(\frac{\pi}{6}\rho\right) \text{ and } \beta_m = 3\beta. \tag{19}$$

From equation (19) one obtains the expected individual particle mass

$$E(M) = \frac{\pi\rho}{6} \exp(3\mu + \tfrac{9}{2}\beta^2). \tag{20}$$

Similarly for the expectation of M^2

$$E(M^2) = \left(\frac{\pi\rho}{6}\right)^2 \exp(6\mu + 18\beta^2). \tag{21}$$

The mass (or volume) weighted diameter distribution is also log-normal with parameters given by equation (22) :

$$D_z \in \mathrm{LN}(\mu + 3\beta^2, \beta^2). \tag{22}$$

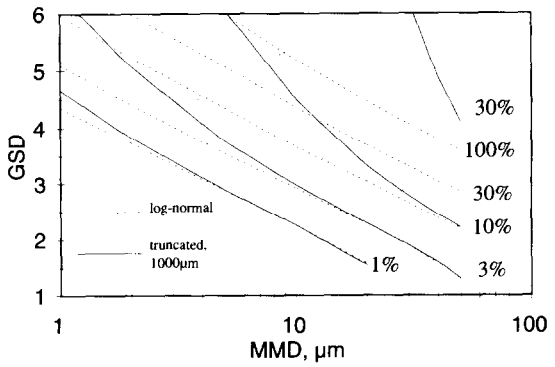

Fig. 1. Lines of equal coefficient of variation of a sample taken from a given volume of air, with expected particle mass 0.1 mg. Log-normal and right truncated (at $d=1000\mu m$) log-normal size distribution. MMD and GSD describe the distribution prior to truncation.

Thus the mass weighted geometric mean diameter, MMD, is

$$\mathrm{MMD}=\exp(\mu+3\beta^2). \tag{23}$$

By combining equations (11), (20) and (21) one gets

$$\mathrm{CV}_{\mathrm{ms}}^2=\frac{1}{\lambda E(V)}\exp(9\beta^2). \tag{24}$$

Notice that this expression depends on λ and β, but not on μ. Expressed in terms of expected total mass of particle sample and mass weighted median diameter, the fundamental coefficient of variation of total particle mass is (using equations (14), (20), (21), and (23))

$$\mathrm{CV}_{\mathrm{ms}}^2=\frac{\pi}{6}\frac{\rho}{E(M_s)}\mathrm{MMD}^3\exp(\tfrac{9}{2}\beta^2). \tag{25}$$

The MMD can be substituted by the mass median aerodynamic diameter MMAD by the relation (particles in the Stokes region, neglecting Cunninghams slip correction) :

$$\mathrm{MMAD}=\sqrt{\frac{\rho}{\kappa}}\mathrm{MMD}, \tag{26}$$

where κ is the dynamic shape factor.

Equation (25) shows that the fundamental coefficient of variation, $\mathrm{CV}_{\mathrm{ms}}$, depends strongly on the size distribution. This is illustrated in Fig. 1, which shows lines of equal $\mathrm{CV}_{\mathrm{ms}}$. The particle population size distribution is given in terms of MMD and GSD. It has been assumed that the expected total mass of aerosol in the sample is 0.1 mg. A mass of 0.1 mg is well above the detection limit of 0.015 mg, which is quoted for occupational hygiene sampling of airborne dusts, using suitable filters (Vaughan *et al.*, 1989). In Fig. 1, MMD ranges from 1 to 50 μm, and GSD ranges from 1 to 6. If equation (26) applies, and if $\rho=1\ \mathrm{g\,cm^{-3}}$, and $\kappa=1$, MMD = MMAD. Then these ranges correspond to those used by Soderholm (1993) for assessing aerosol sampler efficiencies, except that the lower diameter was 0.

DEVIATIONS FROM LOG-NORMALITY

The value of the population parameter $E(W^2)/E(W)^2$, and thus the coefficient of variation will depend on the shape of the size distribution at the extreme upper tail. This is illustrated for $W=M$, by right truncating a log-normal size distribution. Let the non-truncated probability density function for mass be $g(m)$ with parameters given by equation (19). Let the truncation be at $\xi=(\pi/6)\ \rho d_{\max}^3$. The probability density function for the truncated distribution $g^*(m)$ and the jth moment of the truncated distribution is then given

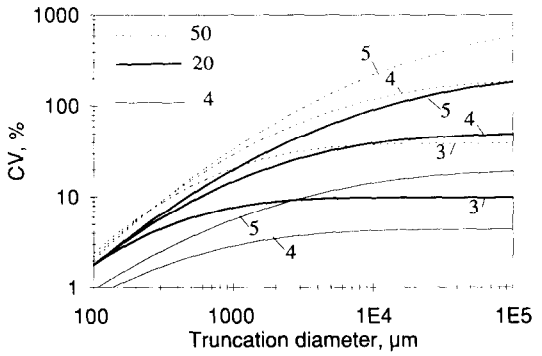

Fig. 2. Fundamental coefficient of variation for some right truncated, log-normal size distributions, as a function of upper diameter truncation. MMD and GSD describe the distribution prior to truncation: MMD = 4, 20, and 50. GSD = 3, 4, and 5.

by the expression:

$$g^*(m)=\frac{g(m)}{\int_0^\xi g(m)\,\mathrm{d}m};\quad E(m^j)=\int_0^\xi m^j g^*(m)\,\mathrm{d}m=\frac{\int_0^\xi m^j g(m)\,\mathrm{d}m}{\int_0^\xi g(m)\,\mathrm{d}m}. \tag{27}$$

Equations (14) and (27) were used to calculate numerically, the values of CV_{ms} for various log-normal size distributions and truncation diameters d_{max}. The result is shown in Fig. 2, for an expected total sample mass of 0.1 mg. Notice that the total sample mass only is a scaling factor, and does not affect the shape of the curves. Figure 2 shows that truncation can have a significant effect, even for particle diameters that are very large compared to the median diameter. MMD and GSD are the size parameters for the log-normal distribution prior to truncation.

In most real situations, very large particles are not part of the airborne particle population. One reason is that their residence time in air is very short so that they cannot be part of the air volume, from which the sample is drawn. If the dust source is a powder there is often an upper particle size limit given by the product specifications. To illustrate the effect of an upper diameter truncation in the particle population from which the sample is drawn, the calculations in Fig. 1 have been repeated using a somewhat arbitrary upper diameter truncation at 1000 μm. This truncation has a considerable effect for large GSD or MMD. Again, MMD and GSD are the size parameters for the log-normal distribution prior to truncation.

To illustrate the effect of deviations from log-normality, as they occur in real aerosols, a real population of particles has also been analyzed. The population consisted of 4321 airborne, man-made mineral fibers, sized by scanning electron microscopy. They were pooled from data available to the authors. The volume equivalent diameter distribution was determined by first calculating volume and then volume equivalent diameters, d, from joint length and diameter measurements, assuming cylindrical shape, then calculating geometric mean and standard deviation of d, see Table 1. The volume distribution is shown in Fig. 3, plotted on a log-probit scale. Suppose first that the diameter distribution is exactly log-normal and has parameters equal to the estimated values $CMD=\exp(\mu_d)=3.67$ μm and $GSD=\exp(\beta_d)=2.58$. Assume also that $\rho=2.5\ \mathrm{g\,cm^{-3}}$. The parameters MMD, GM(M), $E(M)$, and $E(M^2)$ can then be calculated, using equations (19)–(21), and (23). The result is given in Table 1. Similar calculations, but now estimating all parameters directly from the study population, give results also shown in Table 1. They show that deviations from log-normality, which could be judged as minor upon inspecting Fig. 1, have a large influence on these key parameters. As an example the population parameter $E(M^2)/E(M)^2$ differs by a factor of almost 50.

It has to be noted that the calculated value of MMD (Table 1) of the true distribution may be underestimated. This problem has been studied by Mercer (1969/70). Mercer found that the most probable value of median mass weighted diameter, estimated from the diameters of

Table 1. Count median diameter (CMD), geometric standard deviation (GSD), median mass weighted diameter (MMD), mass variability equivalent diameter (MVED), expected mass $E(M)$ of particle, and $E(M^2)/E(M)^2$, calculated for an actual and the fitted log-normal distribution

	Log-normality assumed	Estimated directly from database
CMD (μm)	3.67 (*)	3.67
GSD	2.58 (*)	2.58
MMD (μm)	5.4E + 1	2.4E + 1
MVED (μm)	2.1E + 2	4.2E + 1
GM(M) (ng)	6.5E − 2	6.5E − 2
GSD(M)	1.7E + 1	1.7E + 1
E(M) (ng)	3.6	1.5
$E(M^2)/E(M)^2$	3.1E + 3	6.5E + 1

$\rho = 2.5\ \mathrm{g\,cm^{-3}}$.
(*) From which the other parameters in this column have been calculated.

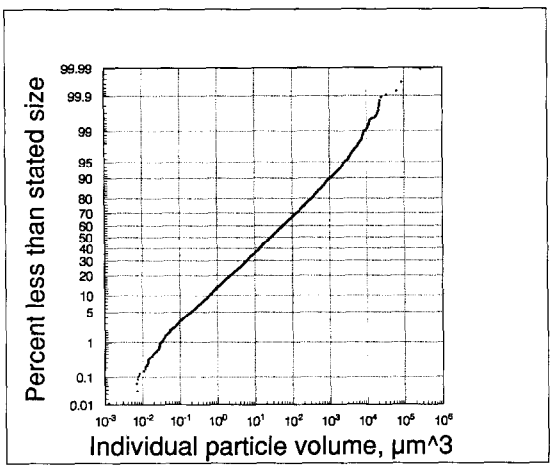

Fig. 3. Cumulative man-made mineral fiber volume distribution on log-probability scale.

N particles drawn at random from a log-normal distribution was less than the true value. For GSD = 2.6, the most probable value was as low as 0.3 times the true value, if $N = 100$ particles had been sized. The most probable measured value was still only 0.7 times the true value for $N = 5000$. This type of bias will also exist for other right skewed distributions. Published size distributions measured with impactors may also be biased due to entry losses in the impactor. Vaughan (1989) has given correction curves for the measured MMD and GSD, assuming a sharp cut-off at 20 μm. Both GSD and in particular MMD will be significantly underestimated, unless corrected for entry losses.

DISCUSSION

The equations for fundamental coefficient of variation can be generalized to other measures than mass. As an example (Appendix), the variability of surface soiling, expressed as percentage of surface area covered by particles is calculated for the log-normal case.

The fundamental coefficient of variation of mass for samples taken from the study population specified in Table 1 is shown in Fig. 4. It is seen that the coefficient of variation is approximately 10% at 0.01 mg. The corresponding expected total number of particles in the sample is 6500. This is a large number, compared with the 100 particles needed for a count coefficient of variation of 10%. If the actual size distribution has the same CMD and GSD

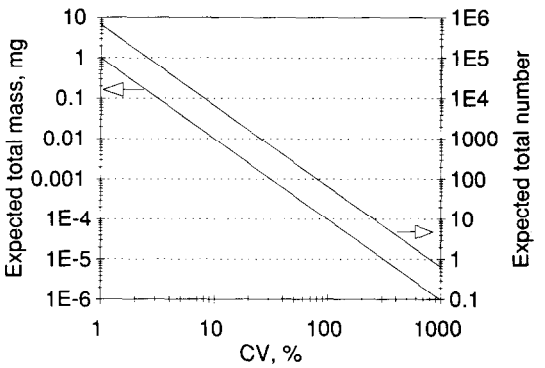

Fig. 4. Expected total particle mass and particle number for given fundamental coefficient of variation of mass. Particle population corresponding to Fig. 3, see also Table 1.

Table 2. Required minimum expected number of particles in a sample to obtain CV_{ms} for total mass less than 25%

$CV_{ms} = 25\%$	GSD = 1.8	GSD = 2	GSD = 2.2	GSD = 2.5
Expected number	360	1210	4300	30600

and was truly log-normal, the required expected total mass would have to be multiplied by 10.7 and the required expected total particle number by a factor 6.9 (as can be calculated using proper parameter values in Table 1).

Table 2 shows the minimum required expected number of particles ($\lambda E(V)$) in a sample if the fundamental coefficient of variation has to be less than 25%. The size distribution is assumed to be log-normal, and thus equation (24) can be used. Table 2 has some implications for, e.g. sizing particles by transmission electron microscopy for total mass determination. It is seen that the time would be poorly spent if all particles of the relevant type were identified and sized, since a very large number would be required to obtain a CV = 25%. Rather one should stratify counting, and "hunt" the few big particles at low magnification. The simpler case, where the stochastic variation of particle number in a drawn sample was neglected, has been studied by Schneider (1993). Numerical simulations were used, and the particle population was assumed to have a trivariate log-normal size distribution. The results were given as percentiles of the distribution of the volume estimator. From the results, the ratio, R, between the 95 and 5% confidence level could be estimated. In one of the given examples, GSD = 2.2 for the volume equivalent diameter distribution. In this case one would have to size 1280 particles, in order to obtain $R < 2$.

Figure 1 shows that experiments involving mass determination of coarse, polydisperse aerosols must be designed carefully. As an example, if differences between aerosol sampling inlets have to be studied by parallel sampling, sufficient mass must be collected to ensure that the experiment has sufficient power to detect a stated difference. The log-normal approximation can serve as a first, conservative estimate. Then the fundamental coefficient of variation of total particle mass may reach 100% for an expected 0.1 mg total particle mass (Fig. 1). For particle size distributions of a more realistic shape at the upper extreme tail, the coefficient may only reach about 30%. Fig. 2, Table 1, and Table 2 demonstrate that one has to assess very carefully how well the modeled size distribution fits the actual size distribution, when calculating CV.

For a monodisperse aerosol, $E(W^2)/E(W)^2 = 1$. The fundamental coefficient of variation CV_{ws}^2 is then the same as for the Poisson distribution

$$CV_{ms}^2 = \frac{1}{\lambda_{tot}} \quad \text{with } \lambda_{tot} = \lambda E(V), \tag{28}$$

λ_{tot} is the expected total particle number in the sample.

For a polydisperse aerosol a similar expression for the coefficient of variation of mass, CV_{ms}, can be obtained. By introducing a mass variability equivalent diameter, MVED, the actual sample of particles can be modeled regarding variability of sampled mass, by replacing the actual aerosol by a monodisperse aerosol with diameters equal to the MVED. The MVED can be calculated in the following way:

The total number of particles with diameter MVED is

$$\lambda_{tot} = \frac{E(M_s)}{\frac{\pi}{6}\rho \mathrm{MVED}^3}. \tag{29}$$

According to the definition of MVED

$$CV_{ms}^2 = \frac{1}{\lambda_{tot}} = \frac{1}{E(M_s)} \frac{E(M^2)}{E(M)}. \tag{30}$$

By combining equations (29) and (30), one obtains

$$\mathrm{MVED}^3 = \frac{6}{\pi\rho} \frac{E(M^2)}{E(M)}. \tag{31}$$

For the log-normal case, MVED is

$$\mathrm{MVED} = \mathrm{MMD} \exp(\tfrac{3}{2}\beta^2) = \mathrm{CMD} \exp(\tfrac{9}{2}\beta^2). \tag{32}$$

The mass variability equivalent diameter gives a "feel" for the coarseness of the aerosol in relation to total mass variability. Some examples are given in Table 3.

Equation (31) can also be used for assessing the coefficient of variation of total determined mass by, e.g. the direct reading instrument for mass, TEOM 1200 Ambient Particulate Monitor (Rupprecht and Patashnick Co., Inc.). This instrument samples at default flow rates, Q, of $3\,\mathrm{l\,min^{-1}}$. If a 5 s read out interval, T, is chosen at a concentration, C, of $10\,\mathrm{mg\,m^{-3}}$, an expected mass, M_s, of 0.004 mg would accumulate over each measuring interval. The expected number of equivalent particles, λ_{tot}, accumulated during interval T is given by the relation

$$\lambda_{tot} = \frac{QCT}{\frac{\pi}{6}\rho \mathrm{MVED}^3} \tag{33}$$

from which

$$CV_{ms}^2 = \frac{\frac{\pi}{6}\rho \mathrm{MVED}^3}{QCT}. \tag{34}$$

Assume that the study population (Fig 3, Table 1) is sampled, using these sampling parameters, and that there are no internal losses. Then MVED = 42 μm, giving $\lambda_{tot} = 26$, and

Table 3. Mass variability equivalent diameter (MVED) in μm, for various log-normal and right truncated at 1000 μm log-normal size distributions

		GSD			
MMD	Truncation	3	4	5	6
4 μm	∞	24	72	192	491
	1000 μm	24	54	84	110
20 μm	∞	122	357	974	2470
	1000 μm	103	161	197	221
50 μm	∞	306	893	2430	6170
	1000∞	201	255	282	297

$CV_{ms} = 20\%$. The mass variability equivalent diameter thus is a useful concept for illustrating the sampled mass variability, using the analogy to the simpler case of sampling particle numbers.

In summary, the variability introduced by the discrete nature of particles must be included when determining overall uncertainty budgets for analytical procedures such as determining aerosol concentrations by the membrane filter method or estimating mass percentage of a particle phase in a bulk sample by microscopy or by chemical analysis. The same has to be done when using direct reading particle mass monitors. The mass variability equivalent diameter is a useful population parameter, since variability can be thought of in terms of number (Poisson) statistics of such equivalent particles.

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APPENDIX

The equations for the log-normal case can be generalized to include any measure of form $W = aD^r$. The distribution of W in terms of the diameter distribution parameters μ and β (see e.g. Aitchison and Brown, 1969) is log-normal, with

$$W \in \mathrm{LN}(\mu_w, \beta_w^2), \tag{35}$$

where

$$\mu_w = \ln(a) + r\mu; \quad \beta_w = r\beta. \tag{36}$$

One then gets for the fundamental coefficient of variation

$$CV_{ws}^2 = \frac{1}{\lambda E(V)} \exp(r^2\beta^2) \tag{37}$$

or

$$CV_{ms}^2 = a\frac{1}{E(M_s)} MMD^3 \exp((-\tfrac{9}{2} + r^2)\beta^2). \tag{38}$$

To illustrate the use of equation (37), the fundamental coefficient of variation of surface soiling is calculated. Let the measure of surface soiling be the fraction, F_s, of surface covered by particles. Since the model assumes that the fraction is small, particle overlap can be neglected. Let the considered sample of the matrix be a delineated area A of the surface, placed at random. Then

$$CV_{ss}^2 = \frac{1}{\lambda E(A)} \exp(4\beta^2) \tag{39}$$

from which

$$CV_{fs}^2 = \frac{\pi/4}{AE(F_s)} \exp(2\mu + 6\beta^2). \tag{40}$$