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RESPIRABLE AEROSOL SAMPLER PERFORMANCE TESTING

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Performance tests for evaluating respirable aerosol sampling methods were developed. The tests entail measurement of the flow-dependent collection efficiency of the aerosol size-discriminating part of the sampler using an aerodynamic particle sizer. The bias relative to an international sampler standard adopted by the American Conference of Governmental Industrial Hygienists, the International Standards Organization, and the Comité Européen de Normalisation is mapped over aerosol size distributions of intended application. Imprecision from flow-effects, filter weighing errors, and intersampler variability is either measured or estimated. Uncertainty in the evaluation experiment itself is explained. Samplers can be rejected if, at specified confidence in the evaluation testing, accuracy is lacking in too large a fraction of samples. Alternatively, the data permit specifying a value of the inaccuracy. Two commercially available personal samplers were subjected to the performance tests suggested. The result was the specification of the flowrates at which the samplers closely match the international definition. Furthermore, upper limits on the inaccuracy at which a sampler can be expected to operate were determined. The effect of the acceptance of such performance criteria rather than sampler design specification for compliance sampling would be to encourage the use of accurate samplers and, thus, the development of improved samplers.

Research into improved aerosol sampler design will become more active within the next few years if sampler performance criteria become established. Until recently, sampling for compliance, as well as most routine industrial hygiene applications in the United States, has relied on a single type of respirable aerosol sampler, the 10-mm nylon cyclone. Sampler design specification has inhibited motivation to develop new samplers, since there has been little chance of a new sampler actually being used.

Since the difficulties in sampling aerosols are many, a sampler-specified system does have advantages. Various unknowns in sampling by differing instruments as well as aerosol size-dependent differences are avoided, if not ignored. However, the sampling system in use today is over 20 years old. Improvements such as modern self-regulating personal sampling pumps, which have developed around gas and vapor sampling, have been slow in coming into aerosol sampling practice. Furthermore, there may be method inaccuracies (from aerosol charge effects, for example), invisible to the user, that would be detected in routine performance testing.

The most recent move towards the establishment of performance standards has been the adoption of an international definition of respirable aerosol by the American Conference of Governmental Industrial Hygienists (ACGIH)[1] for setting TLV®s. The new definition also has been adopted by the International Standards Organization (ISO)[2–3] and the Comité Européen de Normalisation (CEN).[4–5] The need within CEN is the selection of uniform sampling procedures while allowing individual country members choice as to sampling equipment. The new definition is a compromise between the British Medical Research Council (BMRC)[6] version and a more recent proposal of ACGIH,[7] itself one of a series of earlier versions. The CEN and ISO documents further describe sampler performance tests similar to those discussed here.

Within the National Institute for Occupational Safety and Health (NIOSH) and Occupational Safety and Health Administration (OSHA) many sampling and analytical methods have been developed[8–13] with strict accuracy requirements that can be applied, with some modification, to aerosol sampling as well. Specifically, 95% of all samples collected must fall within ±25% of a true or standard value, accounting for both bias and imprecision within the sampling method. Compensation for the inaccuracy in the evaluation experimentation itself also must be made. An additional criterion requires that the mean bias must be less than 10%. Similar is the ISO[14] requirement for aerosol samplers that ". . . under the conditions of use and after application of conversion factors, 67% of results will fall within 10% of the result that would be obtained if the specified curve had been followed exactly." These principles were the basis for aerosol sampler performance criteria proposed earlier[15–16] and provide guidance for this article. Specifically, method bias and imprecision at any aerosol size distribution of interest are computed relative to a standard through the use of models of the sampler's aerosol capture efficiency versus aerodynamic diameter.

Mention of company names or products does not constitute endorsement by the Centers for Disease Control and Prevention.

TABLE I. Variations in Sampler Operation and Evaluation.

Sampler variations:
filter weighing [accounted for externally, since sampler heads are evaluated separately from both filters and sampling pumps]
sampler dimension variation [inter-sampler variability]
mean pump flow uncertainty [accounted for, in part, externally]
particle bounce [not tested here]
pump flow fluctuation [controlled by requiring pulsation dampeners]
wind effects [possibly important at high wind speeds]
sampler loading [not tested here]
aerosol shape and density [assumed negligible]
Evaluation errors:
aerosol concentration drift
aerosol size distribution drift
aerosol count errors
aerosol or sampler charge [controlled in evaluation by aerosol discharge, in application by requiring conductive samplers]
humidity [controlled by using a non-hygroscopic aerosol]
Instrumental error [assumed here under control]:
size calibration
phantom particles
liquid aerosol distortion in aerodynamic particle sizer [no liquid used]
aerosol density effects

The experiments are simplified, however, by separating the errors: personal respirable aerosol sampling systems consist of a sampling head for weighting the smaller aerosol particles in a defined way, together with necessary sampling pump and collection medium. Previous work[16–17] on the 10-mm nylon cyclone has shown that, aside from small intersampler variations arising from inconstant sampler dimensions, the sampling head suffers mainly from systematic error or bias (defined more carefully below); whereas the pump, when not overloaded, and collection medium (filter weighing) generate mainly random errors in assessing respirable mass concentrations. This separation of errors simplifies sampler testing. Namely, the sampling head, heretofore designated as the "sampler," can be tested separately from the filter and pump,[16–18] and the bias in sampling aerosol of any expected size distribution, computed. Verification of the equivalence between computed respirable mass concentrations and measured concentrations using the sampler (including filter holder) in assessing distributed aerosol sizes has been documented.[19]

To this end, an aerodynamic particle sizer, which can measure aerosol size distributions, is used. The sampler's aerosol diameter-dependent collection efficiency is measured as the ratio of the aerosol size distribution after the sampling head to the distribution before the sampling head. The sampler then can be compared to a standard collection efficiency curve such as the new international definition. Specifically, the bias in sampling realistic size distributions is computed.

In addition to bias, variability in the method as applied can be significant. Various sources of imprecision, possibly significant either to sampler use or evaluation, can be classified, as in Table I. The more significant of these effects are accounted for as follows: the sampling efficiency experiment described in the next section measures the intersampler variability (in addition to sampler bias); the effect of filter weighing inaccuracy is estimated from weighing procedures in application; and similarly, the effect of pump flow variability is estimated from the measured flow-rate dependence of the collection efficiency together with knowledge of pumps to be used in practice. Thus a separate imprecision test that would require experimenting on multiple complete sampling units in a dust chamber is eliminated. Such a chamber is difficult to maintain and analyze as to dust homogeneity.

Finally, new means of computing the method accuracy are presented. Details on handling the data from evaluation experiments are given. The approach is then applied in the evaluation of two commercially available personal respirable samplers.

ANALYSIS

Experimental Design

Data was taken under the following conditions:

F [=4] sampler flow rates (Q in L/min): 1.5, 2.0, 2.5, 3.0

S [=8] samplers, numbered s = 1, ... , S

R [=4] replicates, numbered r = 1, ... , R

Sampling heads were evaluated one at a time. Thus a troublesome sampler in-chamber position effect was absent. Also, the lack of filter weighing eliminated another source of evaluation error.

In each of the above 128 experiments, the sampling efficiency E(D) at each of 25 values of aerosol diameter D was

measured. The 128 measured functions of diameter D may be labeled $E_{rs}(D, Q)$. This design was selected since:

(1) flowrate information was needed to optimize flow to best match a standard sampling efficiency $E_{std}(D)$ and to estimate the effect of personal sampling- pump flow uncertainty (described below);

(2) the large number of experiments helped average out the uncertainty arising from variation in the test aerosol concentration during the two measurements whose ratio was needed for each collection efficiency measurement. The evaluation experimental error ($\hat{\sigma}_{eval}$ below) is expected to be negligible; and

(3) low intrasampler head variation was expected.

Modeling Sampling Efficiency Functions

Measured sampling efficiency functions E(D) are modeled by smooth curves, using a standard nonlinear regression routine (such as within SAS®), weighted if necessary to account for inhomogeneity in the variability versus D. There are several reasons for modeling. Counting errors from measurements of a limited number of aerosol particles are averaged out through smoothing among neighboring points in D, if the functions are not over-parameterized. Integration of E(D) × dC/dD (the mass concentration per unit diameter range) for finding the total mass concentration sampled in a distributed dust cloud is simplified. Also, the data are more easily understood in terms of a small number of parameters rather than an array of 25 numbers. Finally, the small number of parameters used simplifies computation of the propagation of errors into other functions.

It is impractical to select a simple model applicable to all samplers likely to be evaluated. However, compounding gaussian noise with simple limiting forms of the sampling efficiency curves has been found to result in useful models.[20] The limiting curves can be very simple. For example, samplers known as "impactors" and "cyclones" are near extensions of the Heaviside step-function $\Theta(D_0 - D)$ (equal to zero at negative arguments, otherwise, one), whereas the "horizontal elutriator" is an extension of the parabola $1 - (D/D_0)^2$. D_0 is a constant parameter closely related to the sampler "cut-size" where the sampling efficiency is 50%.

This approach, though largely empirical, does incorporate what is common to many aerosol samplers, such as the asymptotic conditions:

(1) $\partial E / \partial D \rightarrow 0$; $D \rightarrow 0$ or $D \rightarrow \infty$, and

(2) samplers (e.g., inhalable) often have a tail at large diameter D.

Using a minimum number of parameters, these conditions may be met by compounding noise of variance σ_s^2 onto the above limiting functions:

$$E(D) = \int_0^\infty \frac{dD'}{\sqrt{2\pi}\sigma_s D'} e^{-\frac{\ln^2 D'/D}{2\sigma_s^2}} E_{limit}(D') \qquad (1)$$

where, for example,

$E_{limit}(D) = a\cdot\Theta(D_0 - D)$, cyclones and impactors, and

$$E_{limit}(D) = a\cdot[1 - (D/D_0)^2]\cdot\Theta(D_0 - D), \text{ elutriator,} \qquad (2)$$

allowing intercepts to differ from 1.0 through the parameter a. Within this article a is set equal to one.

The model that results for cyclones and impactors can be expressed in terms of the cumulative normal function Φ as

$$E(D; a, \sigma_s, D_0) = a\Phi\left[\frac{1}{\sigma_s}\ln\left(\frac{D_0}{D}\right)\right] \qquad (3a)$$

The analogous model above for the horizontal elutriator may be expressed as:

$$E(D; a, \sigma_s, D_0) = a\Phi\left[\frac{1}{\sigma_s}\ln\left(\frac{D_0}{D}\right)\right] - a\frac{D^2}{D_0^2}e^{2\sigma_s^2}\Phi\left[\frac{1}{\sigma_s}\ln\left(\frac{D_0}{D}\right) - 2\sigma_s\right] \qquad (3b)$$

For characterizing the flowrate dependence of the collection efficiency, the following model has been found[20] useful and is applied here. Parameters D_0 and σ are modeled as:

$$D_0 = \theta_1(Q/2.0 \text{ L/min})^{-\theta_2}$$
$$e^{\sigma_s} = \theta_3(Q/2.0 \text{ L/min})^{-\theta_4} \qquad (4)$$

$\Theta = \{\Theta_j\}$ are constants, estimated from nonlinear regression of the data. The parameterization of Equation 4 includes shape factors Θ_2 and Θ_4, which adds to the flexibility of the model. Note that the use of 2.0 L/min in Equation 4 is only for convenience and has no significance to the modeling. Parameters modeled by sampler s (= 1, ... , S) are designated by ${}_s\Theta$ (for estimating intersampler variability).

Bias Δ and its Uncertainty

The data from the above experiments result in S model functions $E_s(D) = E(D,Q; {}_s\Theta)$, which can be analyzed as follows. Let dC/dD represent any dust concentration per unit aerosol aerodynamic diameter range (mg/m³/μm) for which the sampler is expected to operate properly. Proposed[4] within CEN is that the range should cover all the log-normal total particle size distributions with geometric standard deviations between 1.75 and 3.5 and mass median diameter < 25 μm. Furthermore, respirable samplers would be evaluated only at aerosol size distributions with the fraction of respirable to total aerosol greater than 5%. This omits sizes beyond the line defined by: (mass median diameter, geometric standard deviation) = (10 μm, 1.5) to (25 μm, 2.75). The narrowest distributions of largest-size aerosols are therefore omitted from consideration. The rationale is that size distributions with small respirable fraction generally either have a small respirable mass concentration (i.e., accuracy in aerosol measurement is not needed except in special cases), or actually consist of small-diameter respirable aerosol mixed with extremely large aerosols, and so would be assessed accurately.

The weighted concentrations $\hat{c}_s$ following the s^{th} sampling head can be estimated at a given size distribution dC/dD:

$$\hat{c}_s = \int_0^\infty dD\ E_s\ dC/dD \qquad (5)$$

The weighted concentration c_{std} collected by a standard or ideal sampler can be estimated in the same way, except that a standard curve $E_{std}(D)$ replaces the measured values $E_s(D)$ in the integrand. Specifically, the international standard sampling efficiency for respirable aerosol (as a fraction of total aerosol) is defined[21] using the log-normal model above:

$$E_{std}(D;\ a,\ \sigma,\ D_0) = \frac{1}{2}[1 + e^{-0.06D}]\ \Phi\left[\frac{1}{\sigma}\ln\left(\frac{D_0}{D}\right)\right], \qquad (6)$$

where $D_0 = 4.25\ \mu m$ and $\sigma = \ln(1.5)$. The factor of Φ in Equation 6 represents the angle-averaged low-wind entrance efficiency into samplers mounted on the body. Denoting $\hat{c}$ as the average over $\hat{c}_s$, the mean bias estimate $\hat{\Delta}$ relative to the standard can then be calculated:

$$\hat{\Delta} = [\hat{c} - c_{std}]/c_{std}. \qquad (7)$$

The uncertainty in this estimate of the bias is an important parameter in the sampler evaluation criteria and is given by:

$$\sigma_{\hat{\Delta}}^2 \equiv \text{var}\ (\hat{\Delta}) = \frac{1}{S}\left[\sigma_{samp}^2 + \sigma_{eval}^2\right] / c_{std}^2 \qquad (8)$$

in terms of constants, σ_{eval} and σ_{samp}. σ_{eval} characterizes errors in the evaluation experiment present randomly at each sampler (intrasampler variation assumed to be negligible in comparison), whereas σ_{samp} gives the intersampler variability. This equation for var($\hat{\Delta}$) takes the place of Equation 2 in Reference 22, with the reference method variance given by the second term. Details on how to estimate σ_{eval} and σ_{samp} and other constants of interest are presented in Appendix A.

Total Imprecision RSD

As mentioned above, earlier work[16] indicates that weighing errors may dominate the sampling method imprecision. However, the proposed performance criterion also takes into account the intersampler variability and the effect of personal pump flowrate inaccuracy in the event that either is large. The following paragraphs discuss intersampler variability and pump effect first, then weighing errors, and finally, the total imprecision of the sampling system.

Intersampler variability, RSD_{samp}

The intersampler variability is determined by the analysis above as the estimate σ_{samp}. Dividing by the standard c_{std} gives the intersampler component of the relative standard deviation as:

$$RSD_{samp} = \sigma_{samp}/c_{std}. \qquad (9)$$

Pump effect, RSD_{flow}

Determining the effect of pump uncertainty requires results at neighboring values of flowrate Q. The mass m sampled at the filter over time t depends on the flowrate Q through:

$$m = Q \cdot t \cdot c(Q), \qquad (10)$$

where the dependence of the weighted concentration c on Q is indicated. Let RSD_{pump} represent the uncertainty in the pump flowrate. Then the effect on the sampled mass m is expressed by RSD_{flow} given approximately by:

$$RSD_{flow} = RSD_{pump}\left|1 + \frac{Q}{c}\frac{\partial c}{\partial Q}\right|. \qquad (11)$$

As proposed within CEN and ISO[2–5], the relative standard deviation of the pump flowrate RSD_{pump} is taken as $3 \cdot RSD_{pump} = 5\%$, specifying the largest flowrate imprecision acceptable.

Weighing Errors, RSD_{weig}

Similarly to RSD_{pump}, the effect of weighing error is estimated by assessing the standard deviation RSD_{weig} in the filter weighing expected in the method as applied. A standard deviation of 40 μg is used here as a specific example of the imprecision in the deposited mass. This translates (for example) to $RSD_{weig} = 4.2\%$ at eight-hour sampling of $c = 1\ mg/m^3$ at $Q = 2$ L/min. All computations in this paper refer to $c_{std} = 1\ mg/m^3$ and the 40 μg value for the weight standard deviation.

RSD

Using the above values, the (total) sampler imprecision is then estimated as:

$$RSD^2 = RSD^2_{weig} + RSD^2_{flow} + RSD^2_{samp}. \qquad (12)$$

Accuracy

A specific definition of accuracy for assessing sampling and analytical methods for industrial hygiene standards compliance decisions is in wide use.[8–13] Suppose that in sampling a true concentration c_{std}, a method gives concentration estimates with mean c and imprecision σ. The accuracy A is defined by an interval that is symmetric about the true value c_{std}; the interval must cover a fraction, designated as α (e.g., 95%), of a method's concentration estimates. Specifically, integrating over all individual concentration estimates c', the equation,

$$\int_{c_{std}(1-A)}^{c_{std}(1+A)} \frac{dc'}{\sqrt{2\pi\sigma}}\ e^{-(c'-c)^2/2\sigma^2} = \alpha\ (\text{e.g, } 95\%), \qquad (13)$$

implicitly defines the accuracy A in terms of mean c, imprecision σ, true c_{std}, and the fraction α. It is convenient to express this in terms of error measures relative to the standard concentration c_{std}. Defining the mean bias Δ and the true relative standard deviation RSD by:

$$\Delta \equiv [c - c_{std}]/c_{std}$$
$$RSD \equiv \sigma/c_{std} \qquad (14)$$

the above equality can be expressed as:

$$\Phi[(\Delta + A)/RSD] - \Phi[(\Delta - A)/RSD] = \alpha,$$

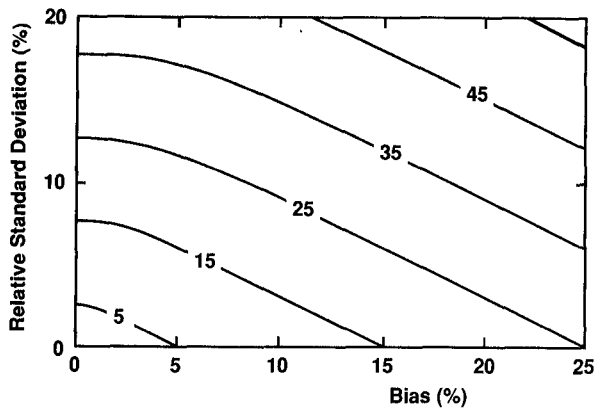

FIGURE 1. Contours showing accuracy A at 95% confidence level in terms of bias and relative standard deviation (RSD)

(15)

where Φ is the cumulative normal function. The function $A(\Delta,RSD)$ can be determined numerically from Equation 15 and is illustrated graphically in Figure 1.

A method evaluation experiment is conducted to estimate the sampling method characteristic quantities, Δ and RSD. The experiment results in estimates, designated as $\hat{\Delta}$ and $\hat{RSD}$. Because of the finiteness of RSD and the number of method replicates and because of variability, designated as σ_{eval} in the evaluation equipment, an evaluation experiment can only imperfectly determine the accuracy A. The design presented here, however, shows how to determine that the accuracy criterion is satisfied with any desired confidence γ (e.g., 95%) in the evaluation experiment.

Several alternatives (integral control, the Bonferroni approach, and tolerance intervals) to computing confidence in the accuracy have been suggested in the literature[22]. Briefly, integral control specifies, for each accuracy A, a one-edged region in $(\hat{\Delta}, \hat{RSD})$ that contains a large fraction γ (e.g., 95%) of measured values $(\hat{\Delta}, \hat{RSD})$. Within Bonferroni control, bias and imprecision are taken to equal their individual upper limits at specified confidences. Finally, the theory[23–24] of linear tolerance intervals has been applied[25] to specify confidence in the evaluation testing. Recently it has been shown[26] that the Bonferroni approach overestimates accuracy confidence limits because of the use of individual confidence limits on bias and imprecision.

An approximate, integral control approach is taken here. The surface A in terms of Δ and RSD_{samp} is approximated as a plane. The result is a rigorous footing for the work of reference 25 and a generalization to account for $\sigma_{eval} > 0$. Details for computing 95% confidence limits ${}_{95\%}A$ on the accuracy are presented in Appendix B.

EXPERIMENT

Procedure

The experimental design and corresponding data analysis given above were tested by experiment on two commercially available respirable aerosol samplers. The Higgins and Dewell (HD) sampler, manufactured in the United States by BGI, Inc. (Model No. BGI-4) was selected because of its successful application in the United Kingdom. The HD sampler is conductive and, as a result, is expected to suffer less from aerosol charge effects than nonconductive plastic samplers. The 10-mm nylon Dorr-Oliver cyclone, manufactured by Mine Safety Appliances Corp. (MSA Part No. 456228), also was tested owing to historical sampling practices in the United States. Four replicate tests of eight sampler examples at four flowrates were performed for each of the two sampler types.

The samplers were tested as isolated samplers facing the wind, one at a time, in a low-speed wind tunnel with cross-section equal to 46 cm × 46 cm. The tunnel was characterized by measuring the air speed over a 25 cm × 25 cm central area in the region where the samplers were to be evaluated. Measurements were accomplished using a hot wire anemometer oriented with flow along the wind tunnel. The anemometer was calibrated using a Kurz Air Velocity Calibration System, Series 400. The result was the mean air speed = 0.54 m/sec. The temporal variation equalled 2.5%. Spatial variation in the air speed over the square grid equalled 3.5%.

Aerosol was introduced at the upstream end of the tunnel by means of a Sonotek ultrasonic nebulizer prior to an X-Static Neutralizer for discharging charged aerosol. The nebulizer aperture and aerosol solution concentration were selected so as to provide a broad distribution of aerosol particles with many present near diameters of rapid change in the cyclones' collection efficiencies. The average aerosol concentration in the tunnel was approximately 5 cm^{-3}.

Aerosol size distributions were determined using a TSI Aerodynamic Particle Sizer 3300 (APS) calibrated with standard polystyrene spheres, Banks Laboratories, Inc. The samplers were connected to the APS by a 1.2 cm (id) copper tube, which made a 180° bend (diameter = 12.7 cm) prior to entry into the APS located beneath the tunnel. The size distribution of aerosol after passing through the sampling head was measured. The collection efficiency was taken then as the ratio of this distribution to the size distribution measured without the sampler. Both distributions traversed the copper tube, and therefore losses within the tube canceled (with and without sampler).

The fixed-diameter copper tubing aperture was normal to the tunnel wind flow, and the flowrate in the tubing was identical to that of the sampler measured. Therefore, the resulting sampling efficiency was measured relative to concentrations as sampled with the specific tubing arrangement, rather than to isokinetic samplers as in the ideal situation. The significance of this set-up can be estimated from the theory of thin-walled samplers.[28] For example, the aspiration efficiency of 4 μm particles at a flow rate equal to 2.0 L/min is calculated to equal 98%.

Constancy of the aerosol in the sampler test region of the wind tunnel was vital. Otherwise, the distribution ratio would be biased from inevitable spatial differences in the positioning of the sampler and isokinetic probe. Spatial homogeneity was achieved by mixing, using turbulence purposely introduced by means of baffles in the tunnel near the aerosol nebulizer and

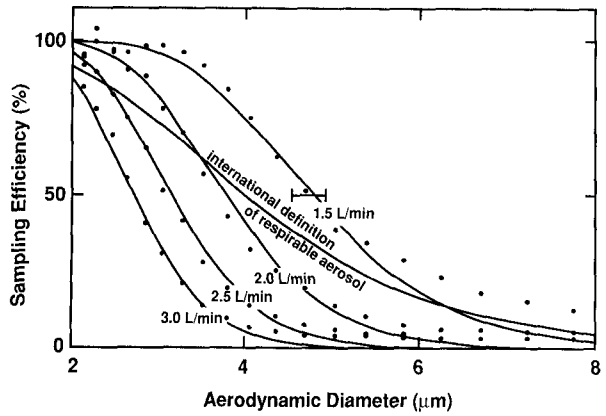

FIGURE 2. 10-mm nylon cyclone collection efficiency data at four flow-rates. Data were averaged over replicates (4) and samplers (8). Solid lines represent model (Equation 4). Error bar represents 4× standard deviation in cut-size, considering intersampler variation

subsequent removal of the turbulence using honeycomb material. The sampling time for measuring each distribution was selected to equal 1 min so as to achieve a balance between counting errors and concentration drift in the wind tunnel. The concentration was measured prior and after each individual sampler evaluation and was found to be very constant: variation equalled 0.5%.

Potassium sodium tartrate was selected as the test aerosol because of its sphericity. The bulk density is equal to 1.77. Electron microscope comparisons with other commonly used test aerosols such as potassium hydrogen phthalate (KHP) indicated that the potassium sodium tartrate particles were spherical and free of pores. Particles were generated with mass median diameter = 4 μm and geometric standard deviation = 2.2. These values were selected to cover the cut-size region of the samplers tested and to minimize spurious counts from excessive numbers of submicrometer particles. The limited number of large particles (> 8 μm) resulted in noise in the measured efficiencies in this region.

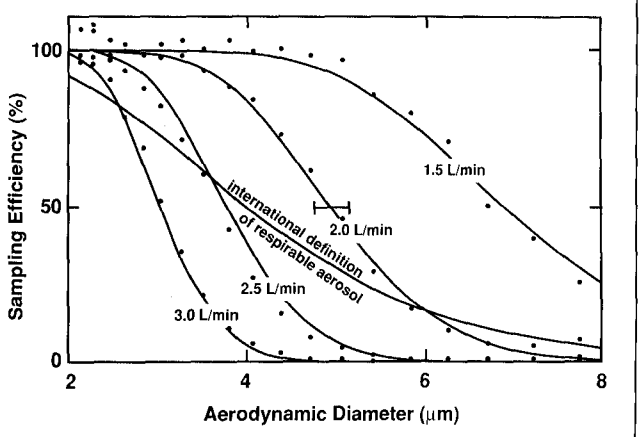

FIGURE 3. HD cyclone collection efficiency data

TABLE II. Fitted Parameters.

HD cyclone:	*10-mm nylon cyclone:*
$\hat{\theta}_1$ = 4.89978 μm	$\hat{\theta}_1$ = 3.75722 μm
$\hat{\theta}_2$ = 1.19682	$\hat{\theta}_2$ = 0.82376
$\hat{\theta}_3$ = 1.23148	$\hat{\theta}_3$ = 1.28863
$\hat{\theta}_4$ = 0.07468	$\hat{\theta}_4$ = 0.01779

Results

The experimental results are given in Figures 2 and 3 for the 10-mm nylon cyclone and the conductive HD cyclone, respectively. The plots reflect the data as averages over the eight samplers and four replicates and show the flow-rate dependent model fits described earlier. The curve-fits were made over aerosol diameters where the noise from low particle count was low: 2 μm < D < 6 μm, for the nylon cyclone and 2 μm < D < 8 μm for the HD cyclone. These regions are large enough to determine cut-sizes and the sampling efficiency gradients over the flow rates tested. To check that the sampling efficiency is low at aerosol diameters larger than these values, a separate, limited experiment was completed using larger test aerosol (mass median diameter = 6 μm and geometric standard deviation = 2.4). The estimated sampling efficiency at 8 μm was found to equal 3% (nylon cyclone) and 2% (HD) cyclone. In the notation of Equation 4 the fit-parameters are listed in Table II.

Equation 6 indicates that the international respirable aerosol cut-size (expressed in terms of total dust) is equal to 4.0 μm (the value 4.25 μm referring to the fraction penetrating the sampler's entrance). The experimental data of Figures 2 and 3 imply that the sampling efficiency of both cyclone types drops towards zero much more sharply than does the international standard. In order to compensate for this effect, optimal cyclone operation was determined by displacement by approximately 0.5 μm relative to the international standard, resulting in 4.5 μm as the cut-size of choice. The data indicate that this cut-size is closely met by the 10-mm nylon cyclone operated at 1.7 L/min and by the HD cyclone with flow rate set equal to 2.2 L/min. Therefore these flow rates were selected for detailed study. Also the flow rate, 2.2 L/min, was studied for the HD cyclone because of results of an independent experiment[27] on the flow rate dependence of the HD sampler manufactured by SIMPEDS for use within the United Kingdom.

Justification for this otherwise arbitrary choice of cut-size is indicated in Figures 4 and 5, where the authors show the estimated mean bias $\bar{\Delta}$, computed as described above (Equation 7) for the two cyclones at the flow rates of interest. As can be seen, the bias of the two sampler types is nearly identical. Furthermore, the bias magnitude remains less than 10% over a wide range of size distributions.

Finally, this bias is combined with errors from filter weighing, flowrate inconstancy (Figures 6 and 7), and intersampler variability (Figure 8 and 9). The result is the method inaccuracy, estimated as in Appendix B. Contour plots of the method inaccuracy are given in Figures 10 and 11.

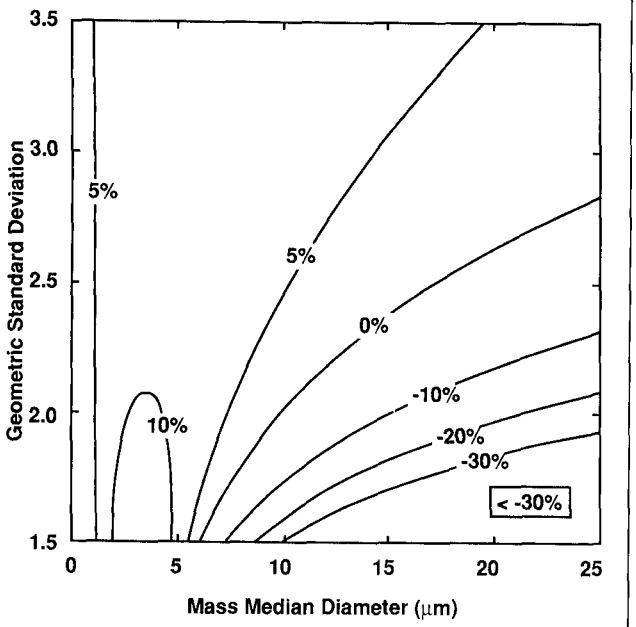

FIGURE 4. Bias map of 10-mm nylon cyclone at 1.7 L/min

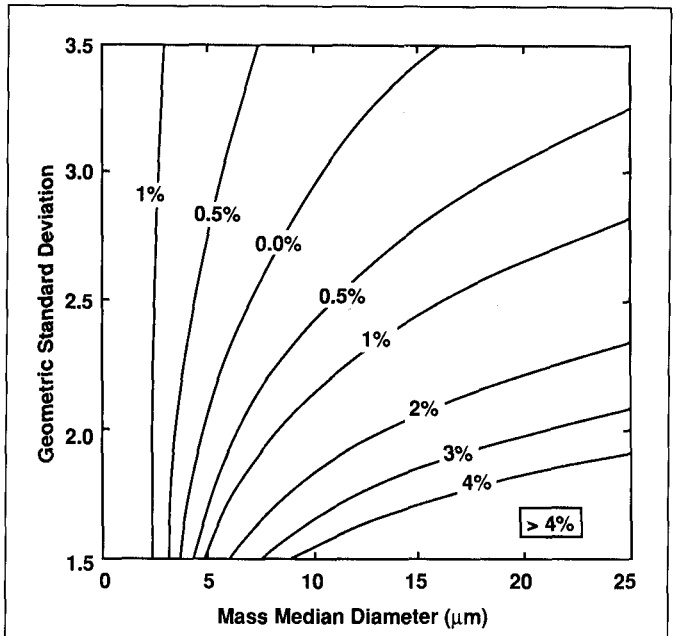

FIGURE 6. Pump induced variability of nylon cyclone

CONCLUSIONS/RECOMMENDATIONS

Two commercially available respirable aerosol samplers were tested for use in a range of aerosol size distributions. Deviation (bias) from the international standard was estimated from the experimental data. Random errors expected in sampler use were in part estimated and in part measured by separating them from the experimental error present in the evaluation testing.

The result is an upper limit on the inaccuracy in 95 out of 100 concentration measurements. This estimated inaccuracy ascribed to a sampling method depends on the confidence, here taken to equal 95%, in the evaluation testing.

The inaccuracy value can be used to demonstrate within sufficient confidence that the concentration is above a specific amount. For example, suppose that a respirable concentration equal to 2.1 mg/m^3 is measured, and that the inaccuracy is less than 37%. Then within the specified confidence limits (95% in the evaluation testing) the probability is at least 95% that the true concentration is greater than 2.1 mg/m^3/ (1.00 + 0.37) = 1.53 mg/m^3.

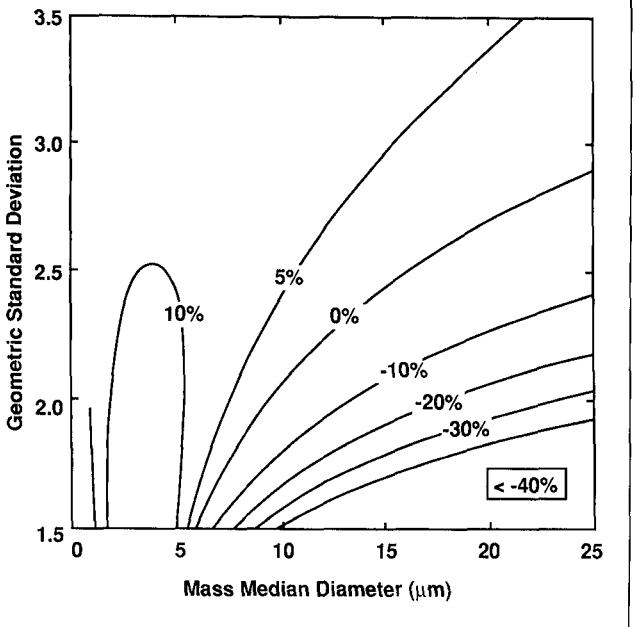

FIGURE 5. Bias map of HD cyclone at 2.2 L/min

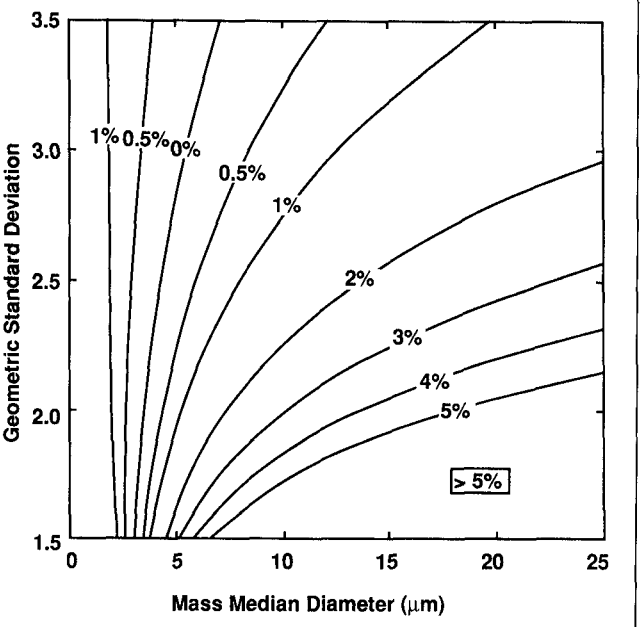

FIGURE 7. Pump induced variability of HD cyclone

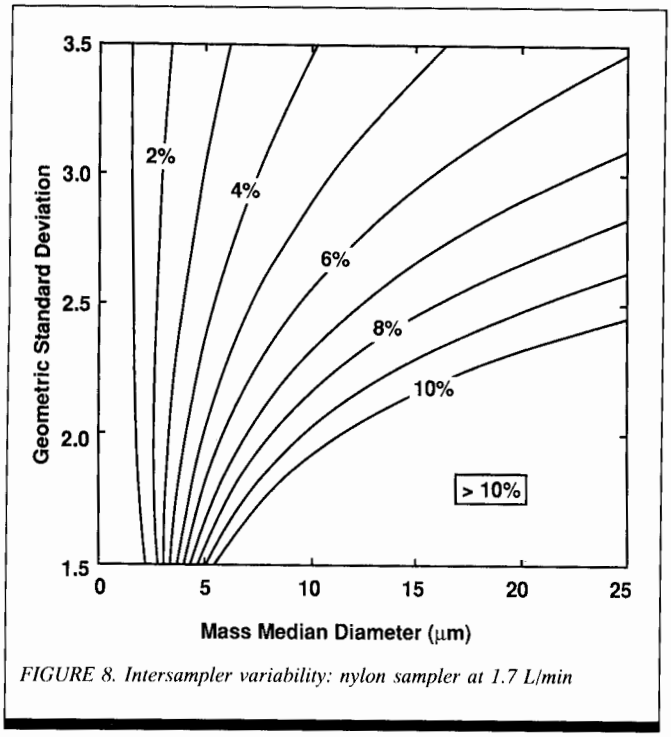

FIGURE 8. Intersampler variability: nylon sampler at 1.7 L/min

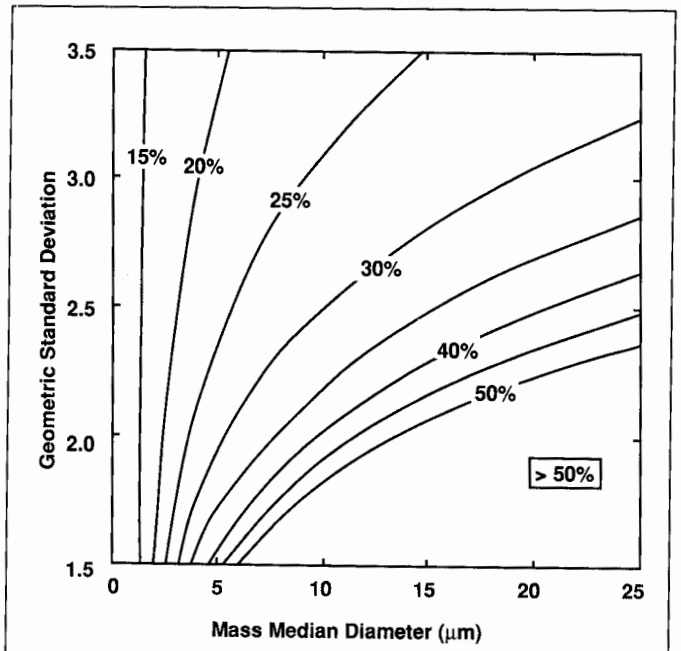

FIGURE 10. Inaccuracy (%) of 10-mm nylon cyclone at 1.7 L/min at the 95% confidence level

Other possibilities for interpreting concentration data exist as well, depending on how the information is to be used. For example, it also can be proved that the concentration is less than a specific value. Furthermore, aside from compliance decisions, there may be instances in which only a multi-day average concentration is needed. In this case the random variations and associated inaccuracy value take on less importance than the bias relative to a standard respirable aerosol.

Again, the evaluation data permit the resolution of questions on established statistical principles. From another point of view, performance standards allow the determination of workplace exposure regardless of the instrument used and environmental parameters. With the actual use of a variety of samplers, instrument designers may see practicality in the development of improved samplers.

As specific examples, improvements may be needed both in the handling of charged aerosol particles[29] and in sampling in windy conditions. Charges on a nonconducting sampler are immobile and therefore provide a localized source of electric

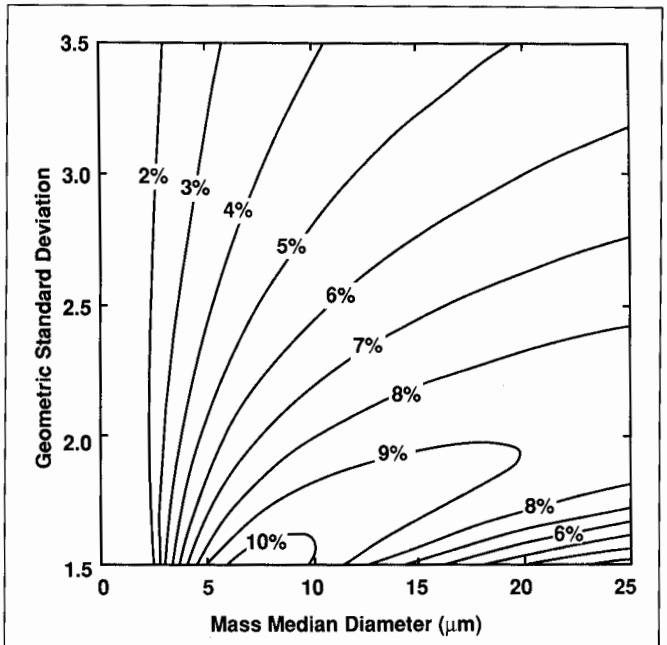

FIGURE 9. Intersampler variability: HD sampler at 2.2 L/min

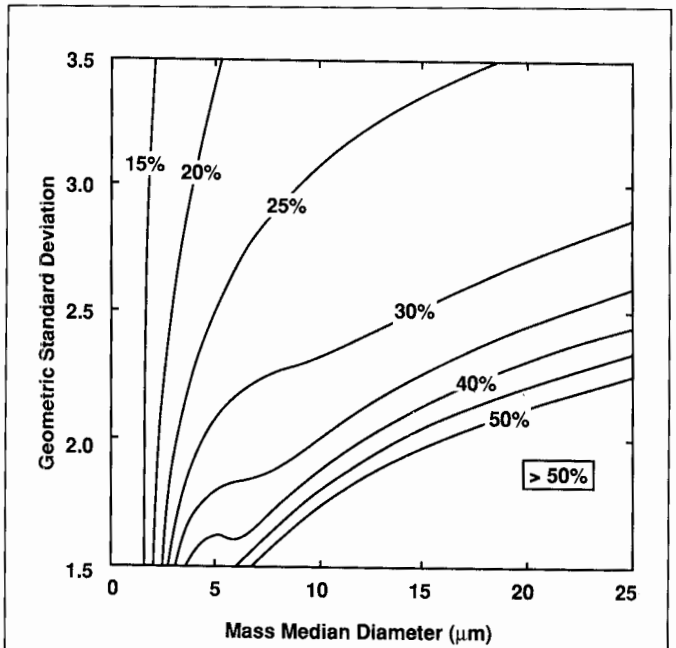

FIGURE 11. Inaccuracy (%) of HD cyclone at 2.2 L/min at the 95% confidence level

field. This can strongly affect the trajectories of charged aerosol particles in the air flowing into the sampler. Quantitatively, a 10% variability has been reported[30] to be associated with charge effects in using the 10 mm nylon cyclone. Furthermore, evidence exists[31] that a charged sampler may undersample moderately charged aerosol by as much as 40%. Finally, the conductivity of the filter holder itself following the 10-mm cyclone may be significant. A 25% increase in the aerosol collected after increasing the holder's conductivity has been reported.[32]

The experiment described in this paper dealt with low wind speed (0.5 m/sec) and turbulence, neither of which is expected to affect the sampling of small-sized respirable particles. Very little information exists on respirable aerosol sampling in higher winds, which can reach 3–4 m/sec in some workplaces. At 3 m/sec a study[33] on the 10-mm nylon cyclone indicates approximately 10% reduction in collection efficiency in sampling aerosol of unspecified size, averaging over all directions of a free-standing cyclone relative to the wind. A similar effect would be expected in personal sampling. In this case the air flow is strongly affected by the body where the sampler is located. The local wind speed (at the sampler) would be reduced when the body faces the wind, but actually could be amplified relative to the ambient wind speed when transverse to the wind.

Other areas of improvement exist as well. For example, orientation-dependent effects may become important at high wind speeds if averaging over directions is not justified. Furthermore, high loadings and the effect of particle "blow-off" should be researched. Also, effects specific to the sampling of liquid aerosol should be addressed.

Despite uncertainties as to the differences between samplers (e.g., HD versus nylon cyclone or human head) under workplace conditions with charge or wind effects, a recommendation as to sampler operation can be made. For best matching of the international definition of respirable dust, the nylon cyclone would be operated at 1.7 L/min and the HD cyclone at 2.2 L/min. Interestingly, the value 1.7 L/min corresponds to an earlier recommendation[34] for sampling according to an Atomic Energy Commission (AEC) respirable dust definition with cut-size equal to 3.5 μm. Data on the cyclone at that time, however, were only preliminary and differ from the present body of data taken by several independent researchers.[15,20,35,36] The AEC definition would be better matched at 2.0 L/min—coincidentally, the traditional sampling rate used by the Mine Safety and Health Administration.

The future undoubtedly will bring further refinements in aerosol sampling. Samplers that provide aerosol size information or at least regional deposition estimates would be useful, particularly in epidemiological research. More novel innovations such as light scattering instrumentation or definition of respirability in terms of particle clearance rates and sub-μm deposition details also are feasible and would require an advance beyond the performance testing applied here. Application of such developments requires the adoption (by the sampler user) of performance standards that allow design flexibility while maintaining accuracy.

APPENDIX A

Statistical Estimation of Needed Parameters

The concentration estimates $\hat{c}_s$ from sampler s are analyzed according to the model

$$\hat{c}_s = c + \varepsilon_{eval\ s} + \varepsilon_s. \qquad (A1)$$

The errors, $\varepsilon_{eval\ s} = N[0,\sigma^2_{eval}]$ and $\varepsilon_s = N[0,\sigma^2_{samp}]$, are represented by their respective standard deviations, σ_{eval} and σ_{samp}. σ_{eval} contains, for example, evaluation concentration fluctuations and aerosol counting errors. σ_{samp} characterizes the inter-sampler variability.

The unknown constant c is the mean weighted concentration that would be approached by averaging many evaluations of many samplers. The constant c is estimated by the sampler average

$$\hat{c} = \frac{1}{S}\sum_s \hat{c}_s. \qquad (A2)$$

Similarly, the variance $\sigma^2_{samp} + \sigma^2_{eval}$ of $\hat{c}_s$ is estimated with $S-1$ degrees of freedom by

$$\hat{\sigma}^2_{samp} + \hat{\sigma}^2_{eval} = \frac{1}{S-1}\sum_s (\hat{c}_s - \hat{c})^2. \qquad (A3)$$

with var($\hat{c}$) given by

$$\mathrm{var}(\hat{c}) = \frac{1}{S}[\sigma^2_{samp} + \sigma^2_{eval}], \qquad (A4)$$

Equation A4 is equivalent to the expression for var($\hat{\Delta}$) given in Equation 8.

σ_{eval} is estimated from the uncertainty in the fitted parameters at fixed sampler s from the (conservative) assumption that all the uncertainty is from experimental error and no part from lack of fit to the model. Then var($\hat{c}_s$) at fixed s is estimated from the nonlinear regression's asymptotic variance-covariance matrix $_s\mathrm{cov}_{ij}$ by

$$\hat{\mathrm{var}}(\hat{c}_s)\big|_s \approx \sum_{i,j} \frac{\partial \hat{c}_s}{\partial \theta_i}\, s\hat{\mathrm{cov}}_{ij}\, \frac{\partial \hat{c}_s}{\partial \theta_j} \quad \text{(fixed s).} \qquad (A5)$$

This quantity is proportional to $(R \cdot F - 4)^{-1}$, where R is the number of replicates and F, the number of flowrates, in the evaluation.

The derivatives, $\frac{\partial \bar{c}_s}{\partial \theta_i}$, can be computed numerically. Averaging over s, an estimate of σ_{eval} is therefore given by

$$\hat{\sigma}^2_{eval} \approx \frac{1}{S}\sum_s \sum_{i,j} \frac{\partial \hat{c}_s}{\partial \theta_i}\, s\hat{\mathrm{cov}}_{ij} \frac{\partial \hat{c}_s}{\partial \theta_j}, \qquad (A6)$$

with approximately $S \cdot (RF - 4)$ degrees of freedom, since $4 \cdot S$ degrees of freedom determine the fitted parameters.

The simultaneous Equations A3 and A6 permit estimation of σ_{samp} and σ_{eval} individually (neglecting lack of fit) for use in Equation A4 and Equation B4 below. As applied to the data presented in this article, the evaluation errors of the experiment described in the text are small:

$$\hat{\sigma}^2_{eval} << \hat{\sigma}^2_{samp}. \qquad (A7)$$

The variability of the estimate $\hat{\sigma}^2_{samp}$ is obtained through the assumed negligible correlation between the right sides of Equations A3 and A6 as in linear regression.

APPENDIX B

Confidence Limit on Accuracy

A confidence limit on the accuracy A, accounting for uncertainty in the evaluation experiment, is computed by approximating the surface, $A(\Delta, RSD_{samp})$, as a plane passing through $(\Delta_0, {}_0RSD_{samp})$, a fixed point determined here by the evaluation experiment itself. Approximating the distribution of general linear functions of bias and RSD_{samp} as normal then leads to a limit on A. The linearization procedure leaves an error dependent on the curvature of the accuracy surface; i.e., an $O(S^{-1} \cdot \sigma^2 A/\partial RSD^2_{samp})$ error in the derived confidence limit. Limiting cases of this approach reduce to the tolerance interval theory developed by Wald and Wolfowitz[23] and applied by Kenny and Lidén[25].

Details are as follows: the accuracy $A(\Delta,RSD)$ (at fixed α, e.g., 95%) may be linearized as the approximation:

$$\begin{aligned} A(\Delta,RSD) &\approx A_0 + \partial A/\partial\Delta|_0 (\Delta - \Delta_0) + \partial A/\partial RSD|_0 (RSD - RSD_0) \\ &= (\partial A/\partial\Delta|_0)\,\Delta + (\partial A/\partial RSD|_0)\,RSD, \end{aligned} \quad \text{(B1)}$$

where the simplification of the second line follows from the fact that A scales with Δ and RSD, as indicated by Equation 15. Linearization of RSD in terms of RSD_{samp} using Equation 12 gives:

$$RSD \approx RSD_0 + ({}_0RSD_{samp}/RSD_0)\,(RSD_{samp} - {}_0RSD_{samp}). \quad \text{(B2)}$$

Therefore, to leading order in υ^{-1}_{rsd},

$$var(\hat{RSD}) \approx ({}_0RSD_{samp}/RSD_0)^2 \cdot \frac{RSD^2_{samp}}{2\upsilon_{rsd}}. \quad \text{(B3)}$$

Furthermore, the variance of the bias estimate is

$$var(\hat{\Delta}) = \frac{RSD^2_{samp}}{S} + RSD^2_{eval}, \quad \text{(B4)}$$

where RSD^2_{eval} represents the second term of Equation 8 and is assumed here to be small enough to produce only perturbative corrections.

Any linear homogeneous combination ζ of RSD_{samp} and bias can be expressed using constants k_1 and k_2 as:

$$\zeta = k_1\,\Delta + k_2\,RSD_{samp}. \quad \text{(B5)}$$

Since $\hat{\Delta}$ and $\hat{RSD}_{samp}$ are independent, the variance of the estimate $\hat{\zeta}$ is given approximately by

$$var(\hat{\zeta}) = k_1^2 \cdot \left[\frac{RSD^2_{samp}}{S} + RSD^2_{eval}\right] + k_2^2 \cdot \frac{RSD^2_{samp}}{2\upsilon_{rsd}}. \quad \text{(B6)}$$

Approximating the distribution of $\hat{\zeta}$ as normal gives an inequality held at the 95% confidence level:

$$\hat{\zeta} > \zeta - 1.64\,RSD_{samp} \cdot \left[\frac{k_1^2}{S} + \frac{k_2^2}{2\upsilon_{rsd}}\right]^{1/2} \cdot \left\{1 + \frac{1}{2}k_1^2 \frac{RSD^2_{eval}}{RSD^2_{samp}}\left[\frac{k_1^2}{S} + \frac{k_2^2}{2\upsilon_{rsd}}\right]^{-1}\right\} \quad \text{(B7)}$$

Now the constants k_1 and k_2 are chosen to make the inequality's right side have the same dependence on RSD_{samp} and bias as A:

$$\begin{aligned} k_1 &= \partial A/\partial\Delta|_0 \\ k_2 - 1.64\left[\frac{k_1^2}{S} + \frac{k_2^2}{2\upsilon_{rsd}}\right]^{1/2} &= \frac{{}_0RSD_{samp}}{RSD_0} \cdot \partial A/\partial RSD|_0. \end{aligned} \quad \text{(B8)}$$

The inequality can finally be rewritten as a 95% confidence limit on A:

$$A < {}_{95\%}A, \quad \text{(B9)}$$

where

$${}_{95\%}A \equiv \hat{A} + 1.64\,\hat{RSD}_{samp} \cdot \left[\frac{k_1^2}{S} + \frac{k_2^2}{2\upsilon_{rsd}}\right]^{1/2} + \frac{1}{2}1.64k_1^2 \frac{RSD^2_{eval}}{\hat{RSD}_{samp}}\left[\frac{k_1^2}{S} + \frac{k_2^2}{2\upsilon_{rsd}}\right]^{-1/2} \quad \text{(B10)}$$

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