Ann. Occup. Hyg., Vol. 57, No. 7, pp. 953–961, 2013

doi:10.1093/annhyg/met005

The Symmetric-Range Accuracy under a One-Way Random Model with Balanced or Unbalanced Data

KALIMUTHU KRISHNAMOORTHY[1] * and THOMAS MATHEW[2]

[1]Department of Mathematics, University of Louisiana, Lafayette, LA 70508, USA; [2]Department of Mathematics and Statistics, University of Maryland, Baltimore, MD 21259, USA

Received 30 September 2012; in final form 22 January 2013; Advance Access publication 2 March 2013

The symmetric-range accuracy A of a sampler is defined as the fractional range, symmetric about the true concentration, that includes a specified proportion of sampler measurements. In this article, we give an explicit expression for A assuming that the sampler measurements follow a one-way random model so as to capture different components of variability, for example, variabilities among and within different laboratories or variabilities among and within exposed workers. We derive an upper confidence limit for A based on the concept of a 'generalized confidence interval'. A convenient approximation is also provided for computing the upper confidence limit. Both balanced and unbalanced data situations are investigated. Monte Carlo evaluation indicates that the proposed upper confidence limit is satisfactory even for small samples. The statistical procedures are illustrated using an example.

Keywords: coverage probability; generalized confidence interval; generalized pivotal quantity; non-central chi-square distribution; upper confidence limit

INTRODUCTION

For quantifying the measurement accuracy of exposure data, the development of accuracy criteria is important. The National Institute of Occupational Safety and Health (NIOSH) accuracy criterion is based on the symmetric-range accuracy A, and the NIOSH accuracy requirement states that a 95% upper confidence limit for A does not exceed 0.25 (see Bartley *et al.*, 2003; Bartley, 2001, 2008; Bartley and Lidén, 2008). By definition, the symmetric-range accuracy A is defined as the fractional range, symmetric about the true concentration C, within which $100(1-\alpha)\%$ of sampler measurements are to be found. Though the NIOSH accuracy criterion applies strictly to intra-laboratory variations, a similar criterion, for example, at the 0.50 level, could be adopted and applied also for controlling inter-laboratory variations. In Bartley (2001) and Bartley *et al.* (2003), an approximation is developed for A, and an approximate 95% upper confidence limit is derived, assuming a normal distribution for the original measurements. In a recent article, Krishnamoorthy and Mathew (2009) have derived an exact expression for A. Also, an accurate upper confidence limit on A may be obtained by exploiting the generalized confidence interval idea. The work of Krishnamoorthy and Mathew (2009) is also in the setup of exposure measurements that follow a normal distribution. It appears that the symmetric-range accuracy and its confidence limit calculation have not been addressed in the context of other models; for example, in the context of a model that involves random effects.

The one-way random model for the log-transformed exposure data is widely used to capture variability among workers when repeated measurements are made on the same worker over time

*Author to whom correspondence should be addressed. Tel: +337 482 5283; fax: +337 482 5346; e-mail: krishna@louisiana.edu

(see Symanski *et al.*, 2006 for a review). The one-way random model has also found applications in the evaluation of sampling/analytical methods where inter-sampler variation is significant. For example, Bartley *et al.* (1994) discuss the 10-mm nylon cyclone whose variation in inner diameter leads to inaccuracy in estimating respirable dust concentrations; a one-way random model is used to capture these variations. The use of a one-way random model results in a situation where a combined uncertainty has to be computed by adding the two variance components (corresponding to variability among and within workers). The calculation of such a combined uncertainty is pointed out in Bartley *et al.* (2003). The same situation arises in inter-laboratory evaluation of methodologies to determine workplace contaminations; the two variance components will now represent the variability between and within laboratories. The goal of this investigation is to derive an exact expression for the symmetric-range accuracy in this scenario and to compute an accurate upper confidence limit, where measurements can be modeled using a one-way random effects model. We have once again used the generalized confidence interval idea in order to derive the required upper confidence limit. We have addressed both balanced and unbalanced data situations. Monte Carlo results are reported to judge the accuracy of the proposed upper confidence limits. We have also illustrated our methodology in both balanced and unbalanced data situations using data from an inter-laboratory evaluation of a methodology to determine trace beryllium in air filter samples.

SYMMETRIC-RANGE ACCURACY

Let X_{ij} denote the j th sampler measurement on the i th worker or from the i th laboratory, assumed to follow the one-way random model given by

$$X_{ij} = \mu + \tau_i + e_{ij},\ j = 1,2,\ldots,n_i, i = 1,2,\ldots,k, \quad (1)$$

where $\mu = E(X_{ij})$ is the overall mean of the measurements, and the τ_i's and e_{ij}'s are independent random variables with $\tau_i \sim N(0,\sigma_\tau^2)$ and $e_{ij} \sim N(0,\sigma_e^2)$. Thus, $X_{ij} \sim N(\mu,\sigma_\tau^2+\sigma_e^2)$. Here, k denotes the number of workers or number of laboratories from where sampler measurements are obtained, and n_i denotes the number of sampler measurements on the i th worker or from the i th laboratory. We note that the X_{ij} represent the original measurements. Furthermore, the random variable τ_i represents an effect due to the i th laboratory or the i th worker.

If C denotes the true concentration and X follows the model in equation (1), then the symmetric-range accuracy A satisfies

$$P((1-A)C < X < (1+A)C) = 1-\alpha, \quad (2)$$

where $0<\alpha<1$, and for NIOSH applications, $\alpha = 0.05$. Here, we have followed the notations and definitions in Bartley *et al.* (2003), Bartley (2001) and Krishnamoorthy and Mathew (2009). As $X \sim N(\mu,\sigma_\tau^2+\sigma_e^2)$, we can write

$$P\big((1-A)C < X < (1+A)C\big) = P\left(\frac{(1-A)C-\mu}{\sqrt{\sigma_\tau^2+\sigma_e^2}} < \frac{X-\mu}{\sqrt{\sigma_\tau^2+\sigma_e^2}} < \frac{(1+A)C-\mu}{\sqrt{\sigma_\tau^2+\sigma_e^2}}\right)$$

$$= P\left(\frac{(1-A)C-\mu}{\sqrt{\sigma_\tau^2+\sigma_e^2}} < Z_0 < \frac{(1+A)C-\mu}{\sqrt{\sigma_\tau^2+\sigma_e^2}}\right), \quad (3)$$

where $Z_0 = (X-\mu)/\sqrt{\sigma_\tau^2+\sigma_e^2}$ follows a standard normal distribution. Let

$$b = \frac{C-\mu}{\sqrt{\sigma_\tau^2+\sigma_e^2}}. \quad (4)$$

Then, equation (3) can be expressed as $P(|Z_0-b| < AC/\sqrt{\sigma_\tau^2+\sigma_e^2})$, or equivalently $P((Z_0-b)^2 < A^2C^2/(\sigma_\tau^2+\sigma_e^2))$. Thus, the symmetric-range accuracy A should satisfy

$$P\left((Z_0-b)^2 < \frac{A^2C^2}{\sigma_\tau^2+\sigma_e^2}\right) = 1-\alpha.$$

Note that $(Z_0-b)^2 \sim \chi_1^2(b^2)$, a non-central chi-square distribution with one degree of freedom and a non-centrality parameter b^2. Hence, $A^2C^2/(\sigma_\tau^2+\sigma_e^2) = \chi^2_{1;1-\alpha}(b^2)$, the $100(1-\alpha)$ percentile of the non-central chi-square distribution. Thus,

$$A = \frac{\sqrt{\sigma_\tau^2+\sigma_e^2}}{C}\sqrt{\chi^2_{1;1-\alpha}(b^2)}, \quad (5)$$

where b is defined in equation (4). Thus, equation (5) gives an explicit expression for A, similar to what is obtained in Krishnamoorthy and Mathew

(2009). Such an explicit expression facilitates the computation of an upper confidence limit for A, as noted below.

Interval estimation with balanced data

Consider the model in equation (1) with balanced data, i.e. the n_i's are equal with n denoting their common value. Thus, we have sampler measurements X_{ij}'s following the model

$$X_{ij} = \mu + \tau_i + e_{ij},\ j = 1,2,\ldots,n, \quad i = 1,2,\ldots,k. \tag{6}$$

Define $\bar{X}_{i\cdot} = \frac{1}{n}\sum_{j=1}^{n} X_{ij}$ and $\bar{X}_{\cdot\cdot} = \frac{1}{kn}\sum_{i=1}^{k}\sum_{j=1}^{n} X_{ij}$. Recall that the τ_i's are random variables representing effects due to the different workers or laboratories. Now let SS_τ denote the between worker or between laboratory sum of squares, and SS_e denote the within worker or within laboratory sum of squares. These are given by

$$SS_\tau = n\sum_{i=1}^{k}(\bar{X}_{i\cdot} - \bar{X}_{\cdot\cdot})^2 \text{ and } SS_e = \sum_{i=1}^{k}\sum_{j=1}^{n}(X_{ij} - \bar{X}_{i\cdot})^2.$$

See Montgomery (2012, Chapter 3). It is known that $\bar{X}_{\cdot\cdot}$, SS_τ, and SS_e are independently distributed with

$$Z = \sqrt{kn}\,\frac{(\bar{X}_{\cdot\cdot} - \mu)}{\sqrt{n\sigma_\tau^2 + \sigma_e^2}} \sim N(0,1),$$
$$\frac{SS_\tau}{n\sigma_\tau^2 + \sigma_e^2} \sim \chi^2_{k-1}, \text{ and } \frac{SS_e}{\sigma_e^2} \sim \chi^2_{k(n-1)}, \tag{7}$$

where χ^2_r denotes the central chi-square distribution with r degrees of freedom (df).

A $100\gamma\,\%$ upper confidence limit for A will now be computed using the generalized confidence interval idea, as done in Krishnamoorthy and Mathew (2009). The first step toward this computation is the derivation of a generalized pivotal quantity (GPQ) for A. As A is a function of μ and $\sigma_\tau^2 + \sigma_e^2$, a GPQ for A can be obtained by substituting the GPQs of these parameters in the expression for A in equation (5). In order to obtain GPQs for the parameters, let $\bar{x}_{\cdot\cdot}$, ss_τ, and ss_e denote the observed values of the random variables $\bar{X}_{\cdot\cdot}$, SS_τ, and SS_e, respectively. The observed values $\bar{x}_{\cdot\cdot}$, ss_τ, and ss_e are numbers computed from a given set of data and are to be treated as fixed. A GPQ is a function of the random variables $\bar{X}_{\cdot\cdot}$, SS_τ, and SS_e, and their observed values $\bar{x}_{\cdot\cdot}$, ss_τ, and ss_e, and is required to satisfy two conditions: (i) given the observed quantities $\bar{x}_{\cdot\cdot}$, ss_τ, and ss_e, the distribution of the GPQ should be free of unknown parameters, and (ii) in the definition of the GPQ, if the random variables $\bar{X}_{\cdot\cdot}$, SS_τ, and SS_e are replaced by the corresponding observed values, the GPQ will simplify to the parameter of interest (the parameter of interest being the symmetric-range accuracy A). To find a GPQ for $\sigma_\tau^2 + \sigma_e^2$, let $\sigma_1^2 = n\sigma_\tau^2 + \sigma_e^2$ and $\sigma_2^2 = \sigma_e^2$ so that

$$\sigma_\tau^2 + \sigma_e^2 = \frac{1}{n}[\sigma_1^2 + (n-1)\sigma_2^2]. \tag{8}$$

A GPQ for σ_1^2 is given by

$$G_{\sigma_1^2} = \frac{\sigma_1^2}{SS_\tau}ss_\tau = \frac{ss_\tau}{\chi^2_{k-1}},$$

and a GPQ for σ_2^2 is given by

$$G_{\sigma_2^2} = \frac{\sigma_2^2}{SS_e}ss_e = \frac{ss_e}{\chi^2_{k(n-1)}}.$$

Substituting these GPQs in equation (8), we obtain a GPQ for $\sigma_\tau^2 + \sigma_e^2$ as

$$G_{\sigma_\tau^2+\sigma_e^2} = \frac{1}{n}\left[\frac{ss_\tau}{\chi^2_{k-1}} + (n-1)\frac{ss_e}{\chi^2_{k(n-1)}}\right]. \tag{9}$$

A GPQ for μ is given by

$$G_\mu = \bar{x}_{\cdot\cdot} - \frac{\sqrt{kn}(\bar{X}_{\cdot\cdot} - \mu)}{\sqrt{SS_\tau}}\sqrt{\frac{ss_\tau}{kn}} = \bar{x}_{\cdot\cdot} - \frac{Z}{\sqrt{\chi^2_{k-1}}}\sqrt{\frac{ss_\tau}{kn}},$$

where Z is defined in equation (7). Finally, a GPQ for A can be obtained by replacing the parameters in equation (5) by their GPQs and is given by

$$G_A = \frac{\sqrt{G_{\sigma_\tau^2+\sigma_e^2}}}{C}\sqrt{\chi^2_{1:1-\alpha}(G_b^2)}, \text{ with } G_{b^2} = \frac{(C - G_\mu)^2}{G_{\sigma_\tau^2+\sigma_e^2}}. \tag{10}$$

An approximation

In order to numerically obtain a percentile of G_A that will provide the required upper confidence

limit, simulation must be carried out a large number of times. Furthermore, the non-central critical value $\chi^2_{1;1-\alpha}(G_b^2)$ must be evaluated for each simulation run and this makes the calculation somewhat time-consuming. The following approximation can be used to avoid the calculation of $\chi^2_{1;1-\alpha}(G_b^2)$ for each simulated G_b^2:

$$\chi^2_{m;p}(\delta) \simeq (m+\delta)\left[z_p\sqrt{\left(\frac{2}{9}\right)\frac{m+2\delta}{(m+\delta)^2}} - \left(\frac{2}{9}\right)\frac{m+2\delta}{(m+\delta)^2} + 1\right]^3 = \chi^{*2}_{m;p}(\delta),$$

where z_p is the $100p$ percentile of a standard normal distribution. The above approximation with $m=1$, $p=1-\alpha$, and $\delta = G_b^2$ can be used to evaluate $\chi^2_{1;1-\alpha}(G_b^2)$ in equation (10) as

$$\chi^2_{1;1-\alpha}(G_b^2) \simeq (1+G_b^2)\left[z_{1-\alpha}\sqrt{\left(\frac{2}{9}\right)\frac{1+2G_b^2}{(1+G_b^2)^2}} - \left(\frac{2}{9}\right)\frac{1+2G_b^2}{(1+G_b^2)^2} + 1\right]^3 = \chi^{*2}_{1;1-\alpha}(G_b^2), \text{ say.} \quad (11)$$

Simulation studies in the sequel indicate that this approximation is very satisfactory and provide results very close to those based on equation (10).

Note that, for given $(\bar{x}_{..}, ss_\tau, ss_e)$, the distribution of G_A does not depend on any unknown parameters, and so the percentiles of G_A can be estimated by Monte Carlo simulation as shown in Algorithm 1. The 100γ percentile of G_A is a 100γ% upper confidence limit for A.

Algorithm 1

1. For given sampler measurements following the model in equation (1), compute $\bar{x}_{..}$, ss_τ, and ss_e.
2. Generate random variates $Z \sim N(0,1)$, χ^2_{k-1}, and $\chi^2_{k(n-1)}$.
3. Compute $G_\mu = \bar{x}_{..} - \frac{Z}{\sqrt{\chi^2_{k-1}}}\sqrt{\frac{ss_\tau}{kn}}$,

   $G_{\sigma_\tau^2+\sigma_e^2} = \frac{1}{n}\left[\frac{ss_\tau}{\chi^2_{k-1}} + (n-1)\frac{ss_e}{\chi^2_{k(n-1)}}\right]$, and

   $G_{b^2} = \frac{(C-G_\mu)^2}{G_{\sigma_\tau^2+\sigma_e^2}}$.
4. Compute $G_A = \frac{\sqrt{G_{\sigma_\tau^2+\sigma_e^2}}}{C}\sqrt{\chi^2_{1;1-\alpha}\left(G_{b^2}\right)}$ or $G_A \simeq \frac{\sqrt{G_{\sigma_\tau^2+\sigma_e^2}}}{C}\sqrt{\chi^{*2}_{1;1-\alpha}\left(G_{b^2}\right)}$, where $\chi^{*2}_{1;1-\alpha}$ is given in equation (11).
5. Repeat Steps 2–4 a large number of times, say, M.

The 100γ percentile of G_A's generated above is a 100γ% upper confidence limit for A.

A special case

If it is assumed that $\mu = C$ (i.e. the bias is negligible) so that $X_{ij} \sim N(C, \sigma_\tau^2+\sigma_e^2)$, then $b=0$ (see equation (4)), and the accuracy range A defined in equation (5) becomes $\frac{\sqrt{\sigma_\tau^2+\sigma_e^2}}{C}\sqrt{\chi^2_{1;1-\alpha}}$, where $\chi^2_{1;1-\alpha}$ denotes the $100(1-\alpha)$th percentile of a central chi-square distribution with one degree of freedom. In this case, the GPQ for A is given by $G_A = \frac{G_{\sigma_\tau^2+\sigma_e^2}}{C}\sqrt{\chi^2_{1;1-\alpha}}$. Clearly, a 100γ% upper confidence limit for A can now be obtained after numerically obtaining the 100γth percentile of $G_{\sigma_\tau^2+\sigma_e^2}$. The estimation of this percentile based on Monte Carlo simulation should be clear from Algorithm 1.

However, we note that in this special case as $A = \frac{\sqrt{\sigma_\tau^2+\sigma_e^2}}{C}\sqrt{\chi^2_{1;1-\alpha}}$, what is required is a 100γ% upper confidence limit for $\sigma_\tau^2+\sigma_e^2$. An approximate upper confidence limit can be explicitly obtained using the Satterthwaite approximation (see Montgomery, 2012, Chapter 13). In order to see this, the chi-square distributions in equation (7) provide us with the unbiased estimators of $n\sigma_\tau^2+\sigma_e^2$ and σ_e^2, say $n\hat{\sigma}_\tau^2+\hat{\sigma}_e^2$ and $\hat{\sigma}_e^2$, given by $n\hat{\sigma}_\tau^2+\hat{\sigma}_e^2 = SS_\tau/(k-1) = MS_\tau$ (say) and $\hat{\sigma}_e^2 = SS_e/[k(n-1)] = MS_e$ (say). From these, we immediately get

$$\hat{\sigma}_\tau^2+\hat{\sigma}_e^2 = \frac{MS_\tau}{n} + \left(1-\frac{1}{n}\right)MS_e,$$

which is a linear combination of MS_τ and MS_e. According to the Satterthwaite approximation, we then have the distribution

$$\frac{\hat{\sigma}_\tau^2+\hat{\sigma}_e^2}{\sigma_\tau^2+\sigma_e^2}\sim\frac{\chi_f^2}{f}, \tag{12}$$

approximately, where the degrees of freedom f is given by

$$f=\frac{\left[\frac{MS_\tau}{n}+\left(1-\frac{1}{n}\right)MS_e\right]^2}{\left[\frac{MS_\tau^2}{n^2(k-1)}+\left(1-\frac{1}{n}\right)^2\frac{MS_e^2}{k(n-1)}\right]}. \tag{13}$$

We note that the degrees of freedom is estimated using the data. A $100\gamma\%$ approximate upper confidence limit for $\sigma_\tau^2+\sigma_e^2$ is now given by $f(\hat{\sigma}_\tau^2\ \hat{\sigma}_e^2)/\chi_{f;1-\gamma}^2$. Consequently, a $100\gamma\%$ approximate upper confidence limit for the symmetric-range accuracy A is now given by

$$\frac{\sqrt{f(\hat{\sigma}_\tau^2\ \hat{\sigma}_e^2)/\chi_{f;1-\gamma}^2}}{C}\sqrt{\chi_{1;1-\alpha}^2},$$

where the degrees of freedom f is given in equation (13).

INTERVAL ESTIMATION WITH UNBALANCED DATA

We shall now consider the model in equation (1) with unbalanced data, i.e. not all the n_i's are equal. Thus, we have sampler measurements X_{ij}'s following the model

$$X_{ij}=\mu\ +\tau_i+e_{ij},\ j=1,2,....,n_i,\ i=1,2,....,k, \tag{14}$$

where the various quantities in the model are as before, satisfying the same distributional assumptions. Note that the number of observations is not the same on the different workers or from the different laboratories. In the case of unbalanced data, a simple approach to compute an upper confidence limit for the symmetric-range accuracy A is to reduce the problem to the case of balanced data using a fairly widely used approximation. In order to introduce the approximation, define

$$h=\frac{1}{k}\sum_{i=1}^{k}n_i^{-1},\ \bar{\bar{X}}=\frac{1}{k}\sum_{i=1}^{k}\bar{X}_{i.}\text{, and } SS_{\bar{x}}=\sum_{i=1}^{k}(\bar{X}_{i.}-\bar{\bar{X}})^2. \tag{15}$$

Then,

$$\bar{\bar{X}}\sim N\left(\mu,\frac{\sigma_\tau^2+h\sigma_e^2}{k}\right).$$

By direct calculation, it can be verified that $E(SS_{\bar{x}})=(k-1)(\sigma_\tau^2+h\sigma_e^2)$. An approximate chi-square distribution associated with $SS_{\bar{x}}$ states that

$$\frac{SS_{\bar{x}}}{\sigma_\tau^2+h\sigma_e^2}\sim\chi_{k-1}^2\ \text{ approximately.} \tag{16}$$

If we define $SS_e=\sum_{i=1}^{k}\sum_{j=1}^{n_i}(X_{ij}-\bar{X}_{i.})^2$, then SS_e is the usual error sum of squares and

$$\frac{SS_e}{\sigma_e^2}\sim\chi_{N-k}^2,\ \text{where } N=\sum_{i=1}^{k}n_i. \tag{17}$$

Using $\bar{\bar{X}}$, $SS_{\bar{x}}$, and SS_e, we shall now imitate the derivation in the balanced case to obtain an upper confidence limit for A. Let $\bar{\bar{x}}$, $ss_{\bar{x}}$, and ss_e, respectively, denote the observed values of $\bar{\bar{X}}$, $SS_{\bar{x}}$, and SS_e, respectively. To find a GPQ for $\sigma_\tau^2+\sigma_e^2$, let $\tilde{\sigma}_1^2=\sigma_\tau^2+h\sigma_e^2$ and $\sigma_2^2=\sigma_e^2$ so that

$$\sigma_\tau^2+\sigma_e^2=\tilde{\sigma}_1^2+(1-h)\sigma_2^2. \tag{18}$$

A GPQ for $\tilde{\sigma}_1^2$ is given by

$$G_{\tilde{\sigma}_1^2}=\frac{\tilde{\sigma}_1^2}{SS_{\bar{x}}}ss_{\bar{x}}=\frac{ss_{\bar{x}}}{\chi_{k-1}^2},$$

and a GPQ for σ_2^2 is given by

$$G_{\sigma_2^2}=\frac{\sigma_2^2}{SS_e}ss_e=\frac{ss_e}{\chi_{N-k}^2}.$$

We note that the GPQ for $\tilde{\sigma}_1^2$ is only approximate as the chi-square distribution associated with $SS_{\bar{x}}$ is only approximate. Substituting these GPQs in equation (18), we obtain a GPQ for $\sigma_\tau^2+\sigma_e^2$ as

$$G_{\sigma_\tau^2+\sigma_e^2} = \frac{SS_{\bar{x}}}{\chi^2_{k-1}} + (1-h)\frac{SS_e}{\chi^2_{N-k}}. \quad (19)$$

A GPQ for μ is given by

$$\begin{aligned} G_\mu &= \bar{\bar{x}} + \frac{\sqrt{k}(\bar{\bar{X}} - \mu)}{\sqrt{SS_{\bar{x}}}}\sqrt{\frac{ss_{\bar{x}}}{k}} \\ &= \bar{\bar{x}} + \frac{Z}{\sqrt{\chi^2_{k-1}}}\sqrt{\frac{ss_{\bar{x}}}{k}}, \end{aligned} \quad (20)$$

where $Z = \frac{\sqrt{k}(\bar{\bar{X}}-\mu)}{\sqrt{\sigma_\tau^2+h\sigma_e^2}} \sim N(0,1)$. Finally, a GPQ for A can be obtained as in equation (10) by replacing the parameters in equation (5) by their GPQs:

$$\begin{aligned} G_A &= \frac{\sqrt{G_{\sigma_\tau^2+\sigma_e^2}}}{C}\sqrt{\chi^2_{1;1-\alpha}(G_b^2)}, \\ \text{with } G_{b^2} &= \frac{(C-G_\mu)^2}{G_{\sigma_\tau^2+\sigma_e^2}}. \end{aligned} \quad (21)$$

The percentiles of G_A can be estimated by Monte Carlo simulation using an algorithm similar to Algorithm 1. The 100γ percentile of G_A is a $100\gamma\%$ upper confidence limit for A. It should be clear that the approximation based on equation (11) that was mentioned for the balanced case can also be used in the unbalanced case. Furthermore, similar to the balanced case, the results simplify in the special case of negligible bias with $\mu = C$. The Satterthwaite approximation can also be used in this special case, as follows. Let $MS_{\bar{x}} = SS_{\bar{x}}/(k-1)$ and $MS_e = SS_e/[k(n-1)]$. In view of equations (16) and (17), an unbiased estimator of $\sigma_\tau^2 + \sigma_e^2$ is given by $\hat{\sigma}_\tau^2 + \hat{\sigma}_e^2 = MS_{\bar{x}} + (1-h)MS_e$. A $100\gamma\%$ upper confidence limit for A, based on the Satterthwaite approximation, is given by

$$\frac{\sqrt{(\hat{\sigma}_\tau^2\ \hat{\sigma}_e^2)/\chi^2_{f;1-\gamma}}}{C}\sqrt{\chi^2_{1;1-\alpha}},$$

where the degrees of freedom f is given by

$$f = \frac{\left[MS_{\bar{x}} + (1-h)MS_e\right]^2}{\left[\frac{MS_{\bar{x}}^2}{k-1} + (1-h)^2\frac{MS_e^2}{N-k}\right]}.$$

We shall conclude our analysis of unbalanced data by making several comments on the chi-square approximation given in equation (16). The approximation is due to Thomas and Hultquist (1978), and these authors have investigated its accuracy. They conclude that the chi-square approximation in equation (16) is accurate except for $\sigma_\tau^2/\sigma_e^2 < .25$, and the data are very unbalanced. Although it is difficult to quantify the extent of the unbalancedness under which the chi-square approximation given in equation (16) becomes unsatisfactory, several authors have used the approximation and have also commented on its accuracy (see e.g. Sahai and Ojeda, 2005, Section 11.8). It should also be noted that in the place of $SS_{\bar{x}}$, there are other formulations of the sum of squares (see once again Sahai and Ojeda, 2005, Chapter 11). Furthermore, these alternative formulations have found applications in industrial hygiene (see Harper *et al.*, 2012). However, for our purpose, $SS_{\bar{x}}$ is preferable due to the chi-square approximation associated with it.

SIMULATION STUDIES

Because a generalized confidence limit is generally known to be only approximate, we note that all of the upper confidence limits derived above are only approximate. In order to assess the accuracy of the upper confidence limits, we performed a simulation study and estimated the coverage probabilities. The simulation was carried out as follows. We first generated 2500 sample statistics $(\bar{x}_{..}, ss_\tau, ss_e)$. For each generated statistic, we used Algorithm 1 with $M = 5000$ to find generalized upper confidence limit for A. The percentage of the 2500 upper confidence limits so obtained that are greater than A is a Monte Carlo estimate of the coverage probability. For the simulation, we chose $1-\alpha = \gamma = 0.95$, and (μ, C) = (2, 2) and (2, 1.5). In other words, we are considering the cases $\mu = C$ and $\mu \neq C$. Our extensive preliminary simulation study indicates that the coverage probabilities are not much affected by the values of n, and they do depend on the values of k. So, simulation results are reported for different

values of k for $n = 2$, 3, and 4. Furthermore, the generalized confidence limits G_A based on Algorithm 1 turned out to be similar to the ones based on the approximation to G_A given in Step 4, and so we used the approximate G_A for our simulation studies. The simulated coverage probabilities are given in Table 1 for the balanced case. It is clear from Table 1 that for smaller k, the generalized upper confidence limits are slightly conservative, especially when $C = \mu$. However, confidence limits are less conservative when C is different from μ.

In the simulation study for the unbalanced case, we choose $k = 6$ and 7, and the results are given in Table 2. The n_i's used in the simulation are given at the end of Table 2. The reported coverage probabilities are close to the nominal level for all the cases considered. Overall, we see that our confidence limits perform satisfactorily in terms of coverage probability, even though they are slightly conservative for smaller values of k.

AN EXAMPLE

The example is on the determination of trace beryllium in air filter samples, and the purpose is an inter-laboratory evaluation of a standardized inductively coupled plasma mass spectrometry method (see Ashley *et al.*, 2009). The relevant data are given in Table 3. We note that there are 20 laboratories, with three replicate observations from each laboratory, except two laboratories, which had only two replicate measurements. We first consider the balanced case after omitting the data for laboratories 13 and 15, which had only two replications each. Thus, $k = 18$ and $n = 3$. Using the notations in the case of balanced data, the summary statistics are

$$\bar{x}_{..} = 8.084,\ ss_\tau = 81.298,\ \text{and}\ ss_e = 33.791.$$

The 95% upper confidence limit based on Algorithm 1 with the exact G_A in Step 4 is calculated as .5329 and with the approximate G_A is .5264. As these upper confidence limits are not <.50, the data do not indicate that the symmetric-range accuracy is <.50. In other words, an accuracy requirement at the .50 level would not be met in the example.

We shall now calculate 95% upper confidence limit for A based on data from all 20 laboratories. Using the notations for the unbalanced case, the required summary statistics are calculated as

$$\bar{\bar{x}} = 8.065,\ ss_{\bar{x}} = 28.329,\ \text{and}\ ss_e = 34.794.$$

Table 1. Coverage probabilities of 95% upper confidence limits for symmetric-range accuracy in the balanced case.

$\mu = C = 1$						$\mu = 1.5$, $C = 2$				
(k, n)						(k, n)				
A	(6,2)	(8,2)	(10,2)	(10,3)	(15,2)	(6,2)	(8,2)	(8,4)	(9,3)	(10,2)
.10	.96	.96	.95	.95	.95	.96	.96	.96	.95	.95
.15	.96	.96	.95	.95	.95	.96	.96	.96	.95	.95
.20	.96	.96	.95	.95	.95	.96	.96	.96	.95	.95
.30	.96	.96	.95	.95	.95	.96	.96	.96	.95	.95
.40	.96	.96	.96	.95	.95	.96	.95	.96	.95	.95

Table 2. Coverage probabilities of 95% upper confidence limits for symmetric-range accuracy in the unbalanced case.

$\mu = C = 1$						$\mu = 1.5$, $C = 2$				
A	(a)	(b)	(c)	(d)	(e)	(a)	(b)	(c)	(d)	(e)
.10	.96	.95	.95	.96	.95	.96	.95	.96	.96	.95
.15	.95	.95	.95	.96	.95	.96	.95	.96	.95	.95
.20	.95	.95	.95	.96	.95	.96	.95	.96	.96	.95
.30	.96	.95	.95	.95	.95	.96	.95	.96	.95	.95
.40	.95	.95	.96	.96	.95	.96	.95	.96	.95	.95

(a)–(e) represent the different choices of the n_i's: (a): (3,2,4,5,3,2); (b): (2,2,2,1,6,12); (c): (4,3,9,2,1,1); (d): (2,2,1,1,3,3,3); (e): (2,2,3,2,4,2,12).

Table 3. Inductively Coupled Plasma-Mass Spectrometry Inter Laboratory Study beryllium results reported by participating laboratories at concentration $C = 10$ μg beryllium.

Lab No.	Replicates		
1	8.75	8.35	8.80
2	9.38	9.56	8.61
3	8.99	7.81	7.16
4	8.60	7.60	6.00
5	8.84	8.80	6.95
6	4.92	5.24	5.87
7	10.1	8.86	8.04
8	9.01	8.36	8.19
9	9.81	9.44	8.16
10	6.42	4.92	4.40
11	10.0	9.60	9.00
12	9.00	7.60	6.50
13	9.28	7.91	
14	9.40	8.82	8.95
15	6.93	7.29	
16	8.90	7.10	4.80
17	7.90	8.33	7.48
18	9.32	9.22	10.1
19	7.89	8.64	6.11
20	9.93	8.55	7.72

Using the expressions in equations (19) and (20), we find

$$G_{\sigma_\tau^2+\sigma_e^2} = \frac{28.329}{\chi^2_{19}} + (1-.35)\frac{34.794}{\chi^2_{38}},$$

and

$$G_\mu = 8.065 + \frac{Z}{\sqrt{\chi^2_{19}}}\sqrt{\frac{28.329}{20}}.$$

Using the above expressions in G_A given in equation (21) and Monte Carlo simulation of 100 000 runs, we estimated the 95th percentiles of G_A as .5186. Thus, the 95% upper confidence limit for A based on all data in Table 3 is .5186. This upper confidence limit is close to the one based only on the balanced data, and so, we arrive at the same earlier conclusion that an inter-laboratory requirement at the 0.50 level would not be met in this example.

An R function routine that calculates upper confidence limits for the symmetric-range accuracy A is posted at www.ucs.louisiana.edu/~kxk4695

DISCUSSION

The computation of confidence limits for some rather complicated parameters comes up in many industrial hygiene applications, and the concept of a generalized confidence interval has proved very fruitful to address such problems. In a series of articles, Krishnamoorthy and Mathew (2002, 2009) and Krishnamoorthy *et al.*, (2006, 2007) have successfully applied the generalized confidence interval idea for the analysis of industrial hygiene data. In particular, Krishnamoorthy and Mathew (2009) have developed an accurate upper confidence limit for the symmetric-range accuracy, using the generalized confidence interval approach, in the context of normally distributed sample measurements. In this article, we have accomplished the same for laboratory measurements that can be modeled using a one-way random model, which is appropriate to model the variability among and within an exposure group or among and within different laboratories. Monte Carlo simulation results have once again demonstrated the accuracy of the proposed methodology. Both balanced and unbalanced data situations are addressed in this work. Given the accuracy of the proposed upper confidence limits, it should be of interest in applications where the one-way random model is appropriate to model the exposure data, and it is of interest to verify accuracy requirements based on an upper confidence limit for the symmetric-range accuracy.

It should be noted that the basic results and approximations used in this article have also been used in Krishnamoorthy and Mathew (2004) and Krishnamoorthy *et al.*, (2007) for the problems involving the computation of tolerance limits in a one-way random model. The generalized confidence interval idea appears to provide a unified approach to address a number of problems of interest in the context of analyzing exposure data.

FUNDING

National Institute of Occupational Safety and Health (R01OH003628).

Acknowledgements—The authors are grateful to Dr David Bartley for several helpful suggestions and for providing the example with the relevant data. The authors also thank Dr Gurumurthy Ramachandran for several comments that resulted in clarification of some of the ideas, and a reviewer for providing useful comments and suggestions.

REFERENCES

Ashley K, Brisson MJ, Howe AM *et al.* (2009) Interlaboratory evaluation of a standardized inductively coupled plasma mass spectrometry method for the determination of trace beryllium in air filter samples. J Occup Environ Hyg; 6: 745–50.

Bartley DL. (2001) Definition and assessment of sampling and analytical accuracy. Ann Occup Hyg; 45: 357–64.

Bartley DL. (2008) The meaning of the bias uncertainty measure. Ann Occup Hyg; 52: 519–25.

Bartley DL, Chen CC, Song R, Fischbach TJ. (1994) Respirable aerosol sampler performance testing. Am Ind Hyg Assoc J; 55: 1036–46.

Bartley D, Lidén G. (2008) Measurement uncertainty. Ann Occup Hyg; 52: 413–7.

Bartley DL, Shulman SA, Schlecht PC. (2003) Measurement uncertainty and NIOSH method accuracy range. In Schlecht PC and O'Connor PF, editors. NIOSH manual of analytical methods. 4th edn. NIOSH Publication No. 2003–154. Washington, DC: US Government Printing Office. pp. 208–28.

Harper M, Lee EG, Slaven JE *et al.* (2012) An inter-laboratory study to determine the effectiveness of procedures for discriminating amphibole asbestos fibers from amphibole cleavage fragments in fiber counting by phase-contrast microscopy. Ann Occup Hyg; 56: 645–59.

Krishnamoorthy K, Mathew T. (2002) Assessing occupational exposure via the one-way random effects model. J Agric Biol Envir S; 7: 440–51.

Krishnamoorthy K, Mathew T. (2004) Generalized confidence limits and one-sided tolerance limits in balanced and unbalanced one-way random models. Technometrics; 46: 44–52.

Krishnamoorthy K, Mathew T. (2009) Inference on the symmetric-range accuracy. Ann Occup Hyg; 53: 167–71.

Krishnamoorthy K, Mathew T, Ramachandran G. (2006) Generalized P-values and confidence intervals: a novel approach for analyzing lognormally distributed exposure data. J Occup Environ Hyg; 3: 642–50.

Krishnamoorthy K, Mathew T, Ramachandran G. (2007) Upper limits for exceedance probabilities under the one-way random effects model. Ann Occup Hyg; 51: 397–406.

Montgomery DC. (2012) Design and analysis of experiments. 8th edn. New York, NY: Wiley.

Sahai H, Ojeda MM. (2005) Analysis of variance for random models: unbalanced data. Boston, MA: Birkhauser.

Symanski E, Maberti S, Chan W. (2006) A meta-analytic approach for characterizing the within-worker and between-worker sources of variation in occupational exposure. Ann Occup Hyg; 50: 343–57.

Thomas JD, Hultquist RA (1978) Interval estimation for the unbalanced case of the one-way random effects model. Ann Stat; 6: 582–7.

Weerahandi S. (1995) Exact statistical methods for data analysis. New York, NY: Springer-Verlag.