Estimation of required monitoring time for obtaining validation data in enclosed spaces

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Methods for estimating airborne contaminant concentrations at specific locations within enclosed spaces, such as mathematical models and computational fluid dynamics (CFD), often are validated against directly measured concentrations. However, concentration variation with time introduces uncertainty into the measured concentration. Failure to determine monitoring time requirements can lead to errors in quantifying representative concentrations, which are likely to be attributed to errors in the method being validated. In the current study, to obtain the representative concentrations at multiple locations with a direct reading instrument, we used the standard deviation ratio (SDR) method to determine the required minimum monitoring time within a specified precision limit. To demonstrate the use of the SDR approach in constructing precision confidence intervals, tracer gas concentrations at nine sampling locations in an experimental room were measured to obtain population parameters. Three flow rates of 0.9, 3.3 and 5.5 m^3 min^{-1} were employed and contaminant concentrations were measured using a photoionization analyser. Monitoring time requirements varied substantially with location within the room and were strongly dependent upon the flow rate of air through the room. The proposed method would be very useful for industrial hygienists and indoor air researchers who sometimes need to obtain several hundred measured concentrations for validation purposes or to perform tests under repeatable conditions in enclosed spaces. This study also showed that the proposed method can be used to devise efficient indoor monitoring strategies.

Introduction

Although tools, such as mathematical models[1–3] and computational fluid dynamic (CFD) methods,[4–13] are available for deterministic assessment of exposure to airborne contaminants in workrooms, most of these methods have not been carefully validated for the wide range of conditions that may be encountered. For example, in order to validate CFD methods for simulating concentrations in a workroom, measurements of contaminant concentrations at many locations throughout the workroom are essential. If a large open area is to be simulated, several hundreds of measurements at multiple points would be required. Implementing such a sampling design with samplers requiring laboratory analysis generally would be prohibitively expensive. Thus, instrumental air monitoring at individual points for short periods is often the only feasible choice.

In a ventilated space containing a contaminant source, obtaining the mean concentration at a fixed location is challenging because the concentration varies with time, even when the air flow rate and contaminant emission rate are held constant.[7,8] Because the means of measured concentrations are used as the standards against which results of other estimation methods are compared, keeping confidence limits of the mean acceptably narrow is essential. The longer monitoring is performed at a location, the more accurate the estimate of the mean concentration. (Here the measurement bias, controlled by appropriate calibration procedures, is assumed to be negligible.) Now, one question arises: "How long should we measure to obtain a representative and precise concentration estimate at a location?" Failure to consider the monitoring time necessary to characterize time-varying concentrations might lead to unacceptable errors in the concentration estimate, and thus, in the validation of the method being tested. Therefore, the purpose of this study was to develop a method for estimating the minimum monitoring time required to estimate the mean concentration of an airborne contaminant in an enclosed space within a specified degree of precision, and to demonstrate the application of this method for an array of sampling points.

Background

The statistical distribution of contaminant concentration at a fixed location in a room is often characterized by the mean and variance. However, when measurements are autocorrelated the mean and variance are not statistically sufficient to characterize the concentration unless the sampling time is long enough so that the effect of autocorrelation is negligible.[14–16] Concentration

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measurements are autocorrelated if paired measurements taken at any fixed time interval have a non-zero correlation coefficient. Autocorrelations can occur when the determinants of concentration, such as effective air exchange rate or contaminant emission rate, vary on time scales longer than the time interval between paired measurements, or if these determinants vary cyclically.

Occupational and indoor measurements are frequently autocorrelated. One method for determining the appropriate sampling time, called standard deviation ratio (SDR) method, was presented by Luoma and Batterman.[17] This approach calculated the required number of sequential samples, and hence the required sampling time. This approach is based upon two parameter estimates: the standard deviation and the autocorrelation. These two parameters are then used to calculate the standard deviation of the mean, also called the standard error, for correlated measurements. The formula for this calculation is given by eqn (1).[18]

$$\sigma_n = \frac{\sigma}{n}\left[n + 2\sum_{i=1}^{n-1} i\rho^{n-i}\right]^{1/2} \quad (1)$$

where σ = standard deviation of the population of all measurements, σ_n = standard deviation of the sample mean of size n, and ρ = autocorrelation coefficient between pairs of measurements separated by one unit of time. It has been known that the first-order autoregressive (Markov) process is the most common in many situations.[17] For the first-order autoregressive process, SDR (σ_n/σ) and ρ are required to find the proper sample size, where the SDR is found by simply taking the ratio of the correlated standard error and the standard deviation. We expanded the SDR method to construct confidence intervals around the mean concentration such that the interval length falls within a desired precision. In this case, a third parameter, the population mean, must also be estimated or measured before using the SDR approach.

In the current study, concentration data at nine locations in an experimental room have been collected to estimate the standard deviation of the population and the autocorrelation coefficients.

Methods

SDR approach to accuracy confidence intervals

The measure of precision (d) used is one-half the width of the confidence interval for μ_c, the true mean concentration (*i.e.*, the distance between the upper/lower confidence limit and the observed mean concentration). For a normally distributed random variable, the confidence interval for the mean is given as:

$$\bar{x} \pm (Z_{1-\alpha/2})\left(\frac{\sigma}{\sqrt{n}}\right) \quad (2)$$

where $\bar{x}$ = sample mean concentration, $\sigma/\sqrt{n}$ = standard deviation of the sample mean (standard error = σ_n), and $Z_{1-\alpha/2}$ = the z-value from the standard normal distribution that relates to the (α/2) percentage. Note that for small sample sizes (generally considered at $n < 30$), the t-distribution with $n - 1$ degrees of freedom should be used instead of the standard normal distribution. Thus, we can define a desired half-width of the confidence interval (d) as:

$$d = (Z_{1\ -\ \alpha/2})\,(\sigma_n)_d \quad (3)$$

where $(\sigma_n)_d$ = standard error corresponding to desired precision, d, and α is such that the interval is a $(1 - \alpha)100\%$ confidence interval.

If a 95% confidence interval is desired, then $Z_{1-\alpha/2} = Z_{0.975} = 1.96$. If the confidence limits are desired to be within 10% of the true mean of measured concentration, then $d = 0.1\ \mu_c$. Then $(\sigma_n)_d$ is calculated by rearranging eqn (3):

$$(\sigma_n)_d = \frac{d}{(Z_{1-\alpha/2})} = \frac{0.1\mu_c}{1.96} = \frac{\mu_c}{19.6} \quad (4)$$

Since the true concentration of the population, μ_c, requires an infinite number of measurements, in practice, this can be replaced with the sample mean, $\bar{x}$, based on a monitoring period long enough to obtain a representative concentration. Thus, eqn (4) can be rearranged as:

$$(\sigma_n)_d = \frac{d}{(Z_{1-\alpha/2})} = \frac{0.1\bar{X}}{1.96} \quad (5)$$

The desired standard deviation ratio, SDR_d, can be calculated as:

$$SDR_d = \frac{(\sigma_n)_d}{\sigma} \quad (6)$$

where σ can be estimated by s (standard deviation of $\bar{x}$), the sample standard deviation of concentration estimated from concentration measurements. Using the value of SDR_d and the value of ρ based on concentration measurements, the necessary sampling size was determined from Fig. 1, an expanded version of a graph presented by Luoma and Batterman;[17] they presented a graph of SDR *versus* sample size with separate lines for values of the autocorrelation coefficient (ρ) from 0 to 0.95. The required monitoring time then was simply calculated by using eqn (7):

$$\text{Required monitoring time} = n_{\text{req}}\ \text{time}_{(x_2-x_1)} \quad (7)$$

where n_{req} = required sample size estimated from the SDR approach, and $\text{time}_{(x_2-x_1)}$ = time interval between two sequential concentration measurements.

The proposed method would be very useful for air researchers who need to have several hundred measurements in an enclosed space to validate CFD methods against experiment results. Another useful application would be for industrial hygienists who often perform repeated measurements in indoor work areas.

Experiment set-up

An experimental room of dimensions (2.86 m(L) × 2.35 m(H) × 2.86 m(W)) was constructed with plywood and insulated with a rigid foam/aluminium foil laminate (Rmax-plus®) in a temperature controlled laboratory, as shown in Fig. 2. The room includes a 1 m high source pedestal, a dilution air inlet, and a room air exhaust to the outside. In order to minimize variation

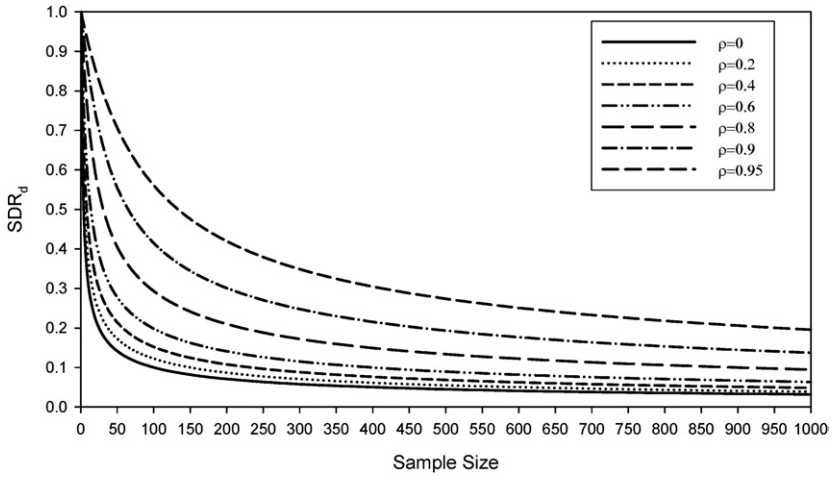

Fig. 1 Predicted variability of the SDR_d (σ_d/σ) for various sample sizes ($n = 1$ to 1000) and autocorrelation coefficients (ρ).

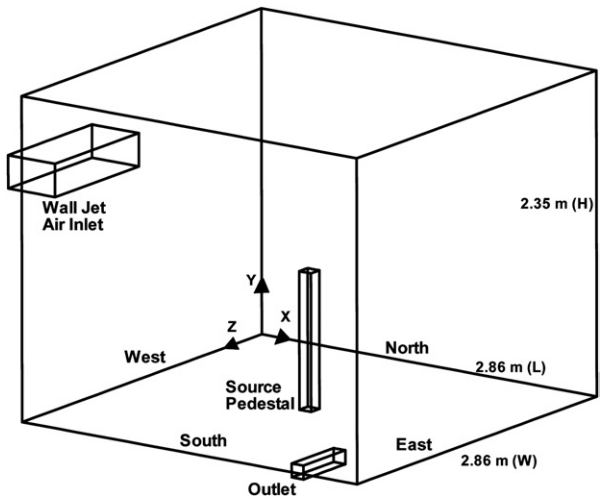

Fig. 2 Experimental room.

of the inlet face velocity across the air inlet, three layers of furnace filter and a flow straightener were placed in the inlet duct. Pure propylene (99.5%) was bled from a compressed gas tank at constant pressure through a calibrated rotameter at known flowrates and entered the room through a 0.1 m diameter, screened opening in the top of the source pedestal. In the current study, pure propylene was selected as the tracer gas because it is relatively non-reactive and non-toxic at the concentration levels used. Only one source location was tested for the simplicity of the experiment and the highly concentrated source was used to generate concentration gradients within the room.

To obtain concentration measurements of the population, three points on the ceiling of the room were selected to represent different room areas. At each point, three Tygon® tubes (ID = 1/16″) were dropped to sample air at three heights above the floor: bottom (0.4 m), middle (1.2 m) and top (2.0 m), resulting in a total of nine monitoring locations. Fig. 3 shows top and front views of the room configuration and sampling points.

Three air flow rates were employed, 0.9, 3.3, and 5.5 m^3 min^{-1}. Those flow rates were determined by measuring velocity along two perpendicular 6 point traverses across the 4 inch diameter, galvanized outlet duct using a thermoanemometer (Model 8350 VelociCalc®, TSI Incorporated, Saint Paul, MN). The velocity profile at the air inlet face was measured to confirm the mass balance between the supply airflow rate and the outlet airflow rate. Air change rate per hour (ACH) for 0.9, 3.3, and 5.5 m^3 min^{-1} were 2.7, 10.2, and 17.2 ACH, respectively. Although the face velocities at the inlet were above the air velocity limits recommended for worker comfort, 0.2 to 0.3 m s^{-1},[4,19] the measured velocity averages at various locations in the occupied zone were less than the comfort limits, even at the highest flow rate (5.5 m^3 min^{-1}) in the current study.[20] Also, the experimental parameters in the current study were set based upon similarity criteria, so that the combination of room size and air exchange rates employed in this study may be used to characterize larger rooms with lower air exchange rates. The use of similarity criteria is described in greater detail elsewhere.[20]

After setting up the desired flow rate for each experiment, the laboratory room air was drawn through the room for approximately 2 h to achieve a steady-state condition. The tracer gas (99.5% propylene) was continuously injected at 150 cm^3 min^{-1} for air flow rates of 0.9 m^3 min^{-1} and 3.3 m^3 min^{-1}, and at 200 cm^3 min^{-1} for 5.5 m^3 min^{-1}. Different tracer emission rates were applied to accommodate the optimal range for measurement with a photoionization (PI) analyser (Model 101, HNu Process Analyzers, Walpole, MA). The expected steady-state room average concentration for each air flow rate would be 166.7, 45.5, and 36.4 ppm, for 0.9, 3.3, and 5.5 m^3 min^{-1}, respectively.

At each monitoring point sequentially, tracer gas concentration measurements were logged every 2 s over a 20 min period (600 measurements), using the PI analyser connected to a data logger (StowAway® Volt; Onset Computer Corp., Pocasset, MA). In the current study, a 20 min period was selected because the measured concentrations at nine sampling points showed consistent patterns of concentrations over that time period, except at the air flow rate of 0.9 m^3 min^{-1}, for which a statistically stationary concentration was not always achieved. (See the Results section for an explanation). The PI analyser was calibrated before and after each test using a known concentration of propylene (100 ppm) in a Tedlar® bag. The inherent instrument

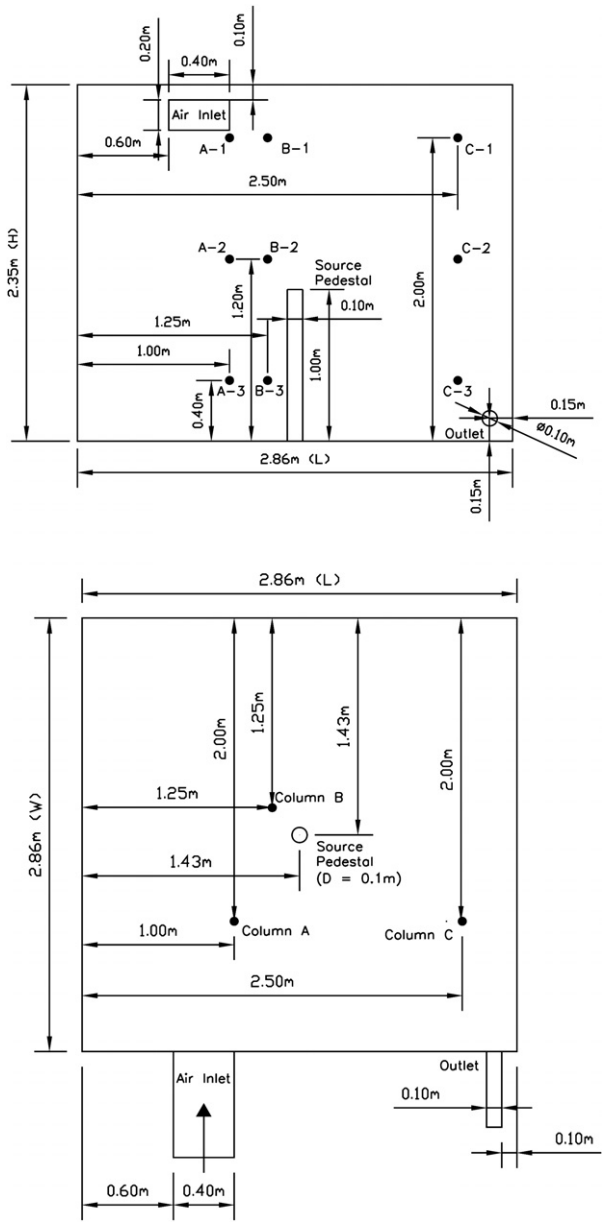

Fig. 3 Sampling locations: front view (top) and top view (bottom).

variability was tested by comparing the coefficient of variation (CV) of concentration at each sampling point in the experimental room with the average CV obtained by monitoring constant gas concentrations in a Tedlar® bag.

An extended study was performed to evaluate the applicability of the theory to contaminant concentrations in the same experimental room. An air flow rate of 5.5 m^3 min^{-1} was used. First, 20 min concentrations were measured from 144 sampling points to estimate concentration of population. After estimating the required monitoring time at each sampling point, measurements were obtained from those points on two different days. At each monitoring location, the ratios of the concentration measured on day 1 to the concentration measured on day 2 was calculated. These ratios were compared across the monitoring locations.

Results and discussion

Concentration monitoring

As shown in Table 1, the highest mean concentrations and standard deviations for two flow rates were observed at the monitoring point B-2, the point located closest to the source. Relative variability of tracer concentrations (CV) differed greatly among monitoring locations. The experimental results indicated that better mixing of room air was observed farther from the source, yielding lower CVs. Higher concentration gradients were observed close to the source. Fig. 4 also shows an example of how measured concentrations varied at two different locations in the room.

The average CV in the Tedlar® bag was 0.047, a substantially lower CV than the CVs for all sampling points, except for three sampling points at an air flow rate of 5.5 m^3 min^{-1}. Thus, the CV of the instrument can be considered negligible because only a small portion of the variability was observed from an artifact of instrument performance.

An interesting preliminary finding at the lowest flow rate (0.9 m^3 min^{-1}) was that statistically stationary concentrations were not reached after 20 min at 6 of the 9 monitoring points. Additional measurements were made at the sampling point C-1 for 5 h, but concentration generally increased over this time period with some short-term variation superimposed; the highest 20 min average instrument reading was about 6.8 times greater than the lowest 20 min average reading. This result indicates an unpredictable air flow movement inside the room due to dominant air motion by thermal convection rather than by forced convention. This result has been discussed in greater detail in elsewhere.[8] Because stationary concentrations were not obtained over extended monitoring periods, the lowest flow rate explored was judged to be impractical for subsequent study. It might require extremely long monitoring times such as days which is impractical to obtain validation data. This unpredictable phenomenon limited the current proposed methodology into a room condition that generates stationary concentrations in the room.

Required sample size by the SDR approach and monitoring time

The autocorrelation coefficient (ρ) was calculated at each sampling location. At most sampling points, the estimated ρ was greater than 0.6, except for one sampling point (A-3 at 3.3 m^3 min^{-1}) in Table 2. After obtaining autocorrelation coefficients, the required sample size using the SDR approach and monitoring time, simply multiplying the required sample size and the time interval of concentration measurements (here, 2 s), were estimated as shown in Table 2. For each condition, three desired precisions, 20% ($d = 0.2$), 10% ($d = 0.1$), and 5% ($d = 0.05$), were employed.

It was observed that the required monitoring time at a constant flow rate varied by more than three orders of magnitude among the nine sampling points. For 3.3 m^3 min^{-1}, the longest monitoring time requirement was observed at point B-2, while for 5.5 m^3 min^{-1}, the longest requirement was observed at point A-1. For 3.3 m^3 min^{-1} at point B-2, more than 6 h and 1.5 h were required to obtain representative concentration estimates for $100d/\mu_c$ values of 5% and 10%, respectively. On the other hand, at some sampling points (A-3, C-1, C-2, C-3) marked

Table 1 Mean, coefficient of variation, and range of concentrations measured every two seconds over a 20 min period

	3.3 m^3 min^{-1}			5.5 m^3 min^{-1}		
	Mean[b]	CV[c]	Range	Mean	CV	Range
A-1	53.0	0.119	36.3–82.2	53.0	0.370	31.7–157.8
A-2	38.4	0.174	24.8–63.1	41.2	0.386	29.8–154.7
A-3	35.5	0.115	26.8–57.4	36.9	0.089	29.8–55.3
B-1	61.8	0.165	42.2–99.6	69.1	0.249	42.3–147.8
B-2	84.3	0.558	34.4–371.6	84.4	0.459	40.4–157.8
B-3	42.6	0.408	28.7–229.7	41.2	0.109	34.2–67.7
C-1	45.8	0.162	28.7–72.7	38.8	0.044	35.4–54.7
C-2	60.5	0.195	40.2–126.4	38.0	0.029	34.2–44.1
C-3	55.0	0.135	40.2–90.1	38.6	0.044	34.2–42.8

The average CV for monitoring constant concentration in a calibration bag: 0.047.[d]

[a] Note: the results for the 0.9 m^3 min^{-1} were not included in the table because stationary concentrations were not reached after 20 min. [b] Average concentration over the time period (ppm). [c] Coefficient of variation (CV = standard deviation/mean). [d] CV of the inherent instrument performance.

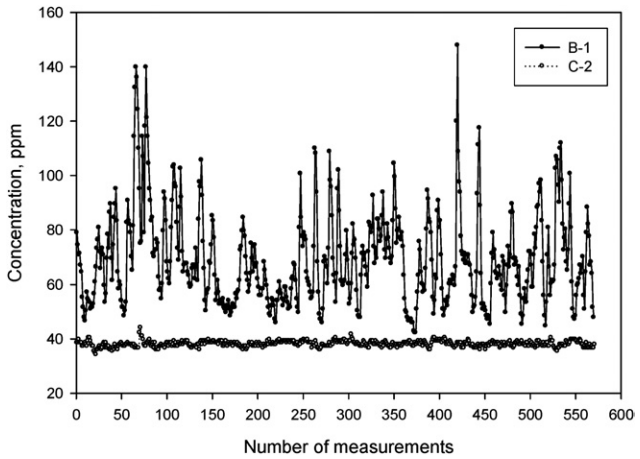

Fig. 4 Measured contaminant concentrations at B-1 and C-2 locations (note: the readings during the first minute (30 measurements) were excluded to allow flushing the sampling line with each sample).

by [e], only one reading (2 s) is necessary. This happened because the variability required to achieve the desired precision is greater than the variability of the measured concentrations ($SDR_d > 1$). Theoretically, the SDR should never exceed one; for example, at the extreme cases, $\rho = 0$ and $\rho = 1$ at all lags, $\sigma_n = \sigma/\mathrm{sqrt}(n)$ and σ, respectively. However, while s, the estimated standard deviation, is dependent upon measured values of concentration, σ_d is independent of these measured values. Thus, when the variability of the measured concentrations is less than the variability required to achieve the desired precision, SDR_d is greater than one. This indicates that as few as one measurement is sufficient to estimate the mean concentration with the desired precision. However, caution should be exercised when basing estimated on a single measurement. For example, in practice to avoid bias, sampling lines should be flushed with a sample from the monitoring point, and adequate time allowed for the instrument to reach a steady-state response before the measurement is taken.

Fig. 5 demonstrates the effect of variability, expressed as CV and ρ on the monitoring time requirement. Both factors are important determinants of required sampling time. The required sampling time increases with increases in either or both of these factors, but the required monitoring time was more sensitive to

Table 2 Autocorrelation coefficients (ρ), required sample size and monitoring time (s)[a]

	3.3 m^3 min^{-1}				5.5 m^3 min^{-1}			
		Required sample size (monitoring time/s)[c]				Required sample size (monitoring time/s)[c]		
Sampling points	ρ[b]	20%[d]	10%[d]	5%[d]	ρ[b]	20%[d]	10%[d]	5%[d]
A-1	0.70	3 (6)	26 (52)	113 (226)	0.89	216 (432)	891 (1782)	3590 (7180)
A-2	0.74	16 (32)	75 (150)	312 (624)	0.86	183 (366)	753 (1506)	3034 (6068)
A-3	0.45	2 (4)	13 (26)	55 (110)	0.93	[e]	69 (138)	329 (658)
B-1	0.82	20 (40)	100 (200)	415 (830)	0.84	62 (124)	268 (536)	1089 (2178)
B-2	0.92	704 (1408)	2855 (5710)	11480 (22960)	0.83	212 (424)	865 (1730)	3477 (6954)
B-3	0.87	224 (448)	919 (1838)	3699 (7398)	0.84	3 (6)	47 (94)	206 (412)
C-1	0.72	11 (22)	58 (116)	242 (484)	0.71	[e]	[e]	13 (26)
C-2	0.84	35 (70)	161 (322)	660 (1320)	0.62	[e]	[e]	2 (4)
C-3	0.78	8 (16)	53 (106)	223 (446)	0.87	[e]	[e]	36 (72)

[a] Note: the results for the 0.9 m^3 min^{-1} were not included in the table because stationary concentrations were not reached after 20 min. [b] Autocorrelation coefficients at lag 1. [c] Monitoring time = required sample size × 2 (s). [d] Desired precision, $100d/\mu_c$. [e] SDR_d is greater than 1; therefore, only a single measurement is necessary, but the sampling tube must be flushed with sample and the instrument allowed to reach steady-state response before this measurement is taken.

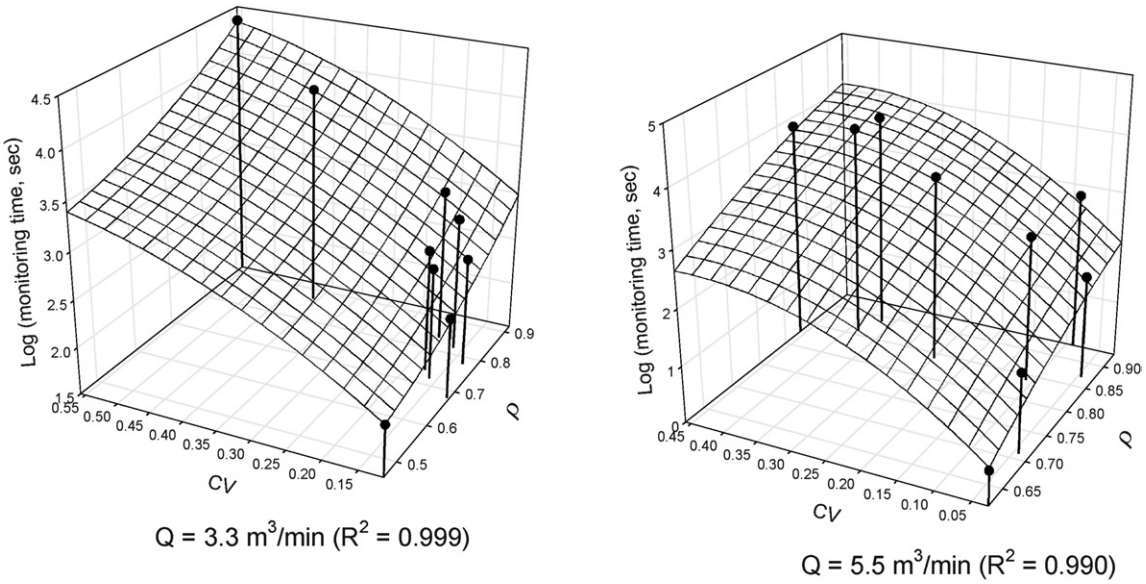

Fig. 5 The log transformed sampling time (in seconds prior to transform) *versus* coefficient of variation (CV) and autocorrelation coefficient (ρ).

the CV than ρ for the data presented here. The least-squares paraboloid surfaces shown in Fig. 5 fit the data well: R^2 values were 0.999 and 0.990 for 3.3 m^3 min^{-1} and 5.5 m^3 min^{-1}, respectively.

Overall, sampling locations nearest to the source produced both the highest CVs and somewhat higher ρ values, and thus required longer monitoring times. On the other hand, less time was required for sampling locations in well-mixed portions of the room, such as the inlet air jet and near the outlet opening.

Application to extended sampling points

For the extended study at 144 points and a flow rate of 5.5 m^3 min^{-1}, the monitoring times estimated by the SDR approach were rounded up to the next highest multiple of 0.5 min.

For a precision limit of 0.1, the required monitoring times ranged from 1 min to 33.5 min. However, for four out of 144 points that required longer than 25 min, a precision limit of 0.15 was applied. On a different day, these times were used to obtain mean concentration values at each point. The ratios of these estimates to the corresponding 20 min mean concentrations were then calculated. As shown in Fig. 6, generally, the distribution of tracer gas shown by the 20 min samples and the estimated concentrations agreed rather well (correlation of coefficients = 0.7839). When one ratio (2.52) was treated as an outlier, the correlation of coefficients was increased to 0.843. The statistical distribution of these ratios was found to be well described by a log normal distribution and the ratio of mean was not significantly different from one (t-test p-value = 0.1676 > α (0.05)). However, for sampling time calculated with a precision limit of 0.1, 30% of the estimated concentrations differed from the corresponding 20 min mean concentrations by more than 10%. Also, 15% of the estimates differed by more than 20% from the 20 min mean. Although the tracer emission rates and the room dilution air flow rates were carefully matched for the two days on which the experiments were performed, based upon agreement of

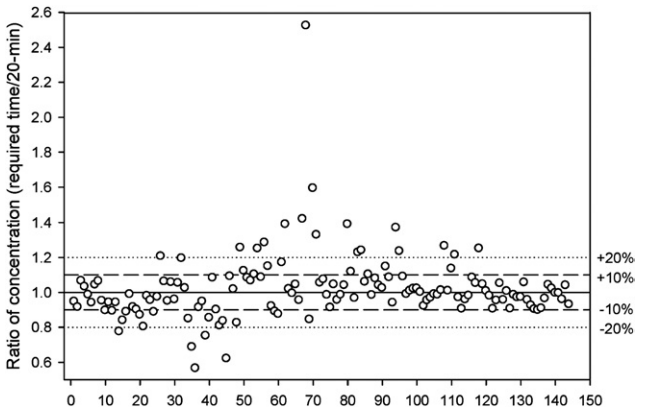

Fig. 6 Estimated tracer gas concentration ratios ($Concen_{(estimated\ monitoring\ time)}/Concen_{(20\ min)}$).

tracer concentration in the exhaust air and the agreement of room mean concentrations, the number of points at which the estimate deviated from the 20 min mean was greater than anticipated. This is likely due to subtle differences in experimental conditions, particularly the wall temperature distribution, which was shown in latter experiments to affect air flow patterns and thus concentration distribution. Thus, it appears the 20 min monitoring data did not fully represent inter-day variability at some monitoring points. This demonstrates that the data used to calculate estimated monitoring time requirements must be representative of the full range of conditions over which subsequent monitoring will be performed, including day-to-day changes.

Conclusions

Validation of innovative methods for estimating contaminant concentration, such as mathematical models or new instruments, against measurements made with a reference method is essential.

However, failure to consider the monitoring time necessary to characterize time-varying concentrations by reference methods may lead to differences between the reference concentration and the concentration estimated by the method being tested. Such differences are likely to be attributed to errors in the tested method, possibly leading to unwarranted rejection of new methods.

In the current study, the implementation of the SDR approach was expanded to calculate required sampling time in a steady-state experimental room, particularly in order to calculate confidence intervals of a desired precision around the mean concentration. Supply air flow rates and source emission rates were kept constant during experiments.

At the lowest air flow rate studied ($Q = 0.9$ m^3 min^{-1}), statistically stationary concentrations were not reached, even after five hours of monitoring at one sampling point. At a randomly selected sampling point, the ratio of instrument readings between the highest 20 min and the lowest 20 min was approximately 6.8. Thus, adequate monitoring times for this condition could not be estimated. These air flow conditions are not extreme and thus similar circumstances may be encountered elsewhere. The implication for developing monitoring strategies is that accurate characterization of mean long-term concentrations in rooms with low air flow rates may require extremely long monitoring times, even when emission rate and air flow rate are relatively constant.

This study also showed that monitoring time requirements vary substantially with location within a room and depend on airflow rates, which cause variation in airflow patterns and thus, in concentration. For the other two higher flow rates, the required monitoring times varied by more than three orders of magnitude among the nine sampling points. Locations near the air inlet and the outlet openings showed the least variation and required relatively short monitoring times, whereas large random variations were observed near the source, requiring longer monitoring times.

The proposed method would be very practical to industrial hygienists and indoor air researchers who sometimes need to obtain several hundred measured concentrations for validation purposes or to perform tests under repeatable conditions in enclosed spaces.

An important limitation in using the SDR method is that the preliminary data used for determining monitoring time must be representative of the full range of conditions for which subsequent monitoring will be performed. Thus, the impact of any changes in air flow, emissions, and room convection patterns or other factors affecting the statistical features of concentration may lead to underestimation of monitoring time requirements.

Disclaimer

The findings and conclusions in this report are those of the authors and do not necessarily represent the official position of the Centers for Disease Control and Prevention.

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