WHY CHARCOAL DEVICES MUST BE ANALYZED SOON AFTER MEASUREMENT: UNCERTAINTY AND MINIMUM DETECTABLE CONCENTRATION

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Abstract

The value of a radon measurement is meaningless unless one knows two additional pieces of information about the measurement; the total uncertainty and the Minimum Detectable Concentration (MDC). These statistics are particularly important when making measurements with devices that capture radon and then are analyzed later; such as grab scintillation cells and charcoal devices. During the delay between sampling and analysis, the quantity of radon in the device constantly decreases due to its decay with a half-life of about 3.8 days. Using 4-inch charcoal canisters as an example, and using typical values of analysis parameters, it is shown that the total uncertainty and the MDC for the measurement both increase with time after the sampling period and can become unacceptable, thus rendering the measurement useless. This illustrates the importance of analyzing these devices as quickly as possible after sampling.

Introduction

Most people realize that charcoal devices capture radon which then decays with a half-life of about 3.8 days, and therefore the device must be analyzed soon after the measurement is made. However, many do not understand the full implications; specifically, the effect of delay time on measurement uncertainty and the Minimum Detectable Concentration (MDC). It would be helpful to realize that the measurement uncertainty is related to the ratio of the "signal" from the charcoal to the "noise" which is the background of the analysis system. With decreasing radon activity in the charcoal, this signal-to-noise ratio becomes smaller, and it becomes difficult to discern the difference between signal and noise. The signal-to-noise ratio is highest immediately following the exposure of the charcoal. Examples are given here, using typical values of pertinent parameters for 4-inch open-face charcoal canisters, to demonstrate the counting uncertainty at the 2-sigma level and the MDC as a function of delay time.

Method

The equation originally published by George (1984) has been used by many for calculating the radon concentration from charcoal canisters. That equation is as follows:

$$C = NCR / (CF * t_e * \varepsilon * DF) \quad (1)$$

where C = Rn concentration (pCi/L)
NCR = net count rate (cpm)
CF = calibration factor (L/min)
t_e = exposure time (min)

ε = counting efficiency (cpm/pCi)
DF = decay factor (unitless)

The decay factor, DF, corrects for the decay of radon from the midpoint of the measurement duration to the beginning of the analysis. The counting efficiency, ε, is determined by counting a standard canister of the same geometry containing radon in equilibrium with a known activity of radium-226.

Minimum Detectable Concentration

The Lower Limit of Detection (LLD) is the lowest net count rate (cpm) that is statistically greater than background. The LLD is a function of the background count rate of the analysis system and the counting times for the sample and for the background. The LLD at the 95% confidence level is calculated using the following equation (Currie, 1968):

$$LLD = 2.71/t_s + 3.29\ (R_b/t_b + R_b/t_s)^{1/2} \quad (2)$$

where LLD = lower limit of detection (cpm)
R_b = background count rate (cpm)
t_b = background counting time (min)
t_s = sample counting time (min)

The MDC (pCi/L) is calculated by dividing the LLD by the same calibration factor, or combination of factors, that is used to convert the sample net count rate to radon concentration. For these examples, the LLD is substituted for NCR in equation (1):

$$MDC = LLD / (CF * t_e * \varepsilon * DF) \quad (3)$$

Counting Uncertainty

The total uncertainty of the measured radon concentration is a function of the individual uncertainties of all the terms on the right-hand side of equation 1. Only the uncertainty associated with the net count rate (NCR), hereafter called the "counting uncertainty," is addressed in this paper, because NCR is the only term in equation (1) whose uncertainty increases with delay time. However, it should be realized that this is only one component, and perhaps not the largest component, of the total uncertainty.

The counting uncertainty S_{CT} at the 95% confidence level expressed as a percentage of the net count rate is calculated using the following equation:

$$S_{CT} = 200 * (R_s/t_s + R_b/t_b)^{1/2} / NCR \quad (4)$$

where S_{CT} = counting uncertainty at 95% confidence level (%)
R_s = sample gross count rate (cpm)
200 = factor consisting of 2 for the 95% confidence level and 100 to convert from fraction to percentage

All other terms are as defined above.

This equation is based on Poisson "counting statistics," which is described in numerous texts on radiological sciences or health physics (for example, Cember & Johnson, 2008).

A value of NCR can be calculated for any assumed value of radon concentration using equation (1) rearranged as follows:

$$NCR = C * CF * t_e * \varepsilon * DF \quad (5)$$

The sample gross count rate, R_s, can be calculated by adding NCR and R_b. Using typical values for the various parameters for 4-inch open-face charcoal canisters, and typical values for the measurement system, a value of S_{CT} can be calculated for any assumed value of radon concentration, C, using equations 4 & 5.

Results and Discussion

Effect of Background Count Rate

The values listed in Table 1 for several parameters for 4-inch open-face charcoal canisters were used with equations 2 & 3 to calculate the MDC for values of 0 to 6 days for the delay from the end of the exposure period to the analysis. Three values of background count rate were used to demonstrate how the MDC changes with that parameter. The value for the calibration factor, CF, is typical for a two-day exposure and a relative humidity of about 50%. The results of the calculations are shown in Figure 1.

Table 1. Parameter values used for demonstrating effect of background count rate on the MDC

Parameter	Value
Exposure time, t_e	48 hours
Counting efficiency, ε	0.39 cpm/pCi
Calibration factor, CF	0.096 L/min
Sample counting time, t_s	10 min
Background counting time, t_b	10 min
Background count rate, cpm	100, 200 & 300 cpm

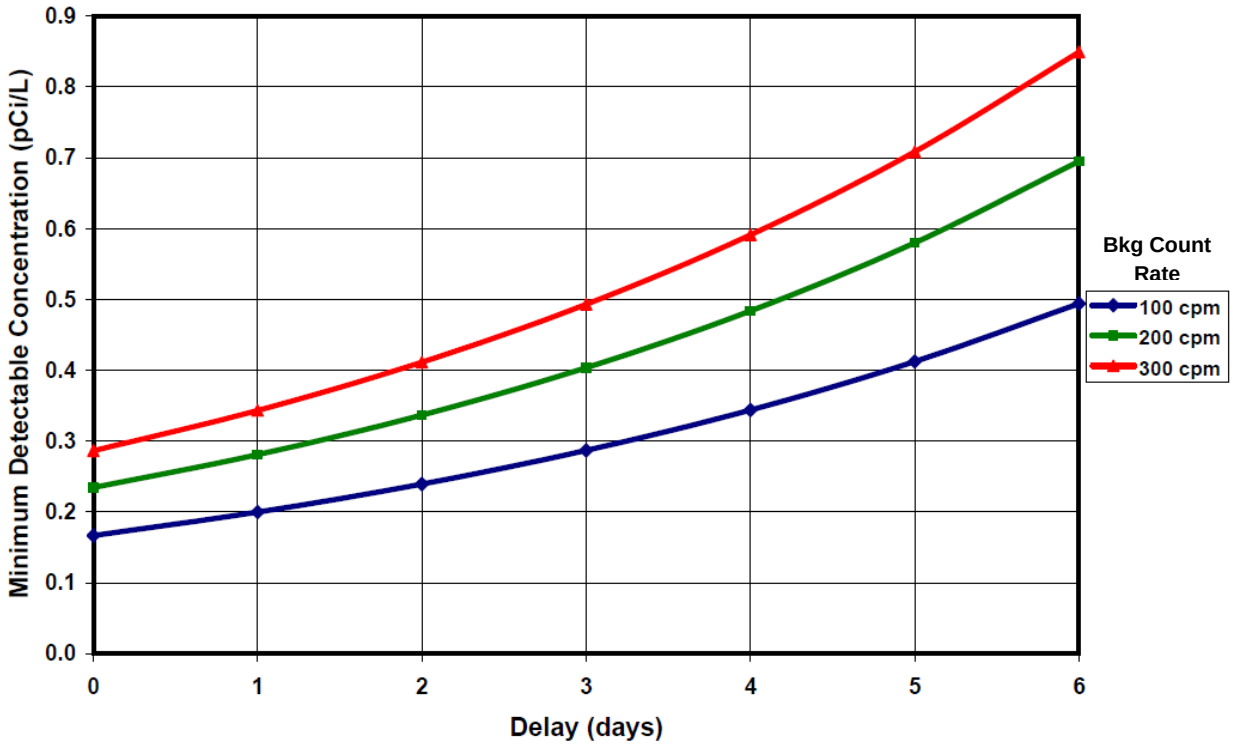

Figure 1. Effect of background count rate on MDC

The LLD calculated using equation 2 does not change with delay time, but because of the factors used to convert LLD to MDC in equation 3, the MDC increases with delay time. The values of background count rate are in the range that is typical of sodium iodide gamma spectroscopy systems used to analyze charcoal canisters.

The values in Table 1 were also used to calculate the counting uncertainty, S_{CT}, using equations 4 & 5. Unlike the MDC, S_{CT} is a function of the radon concentration. A value of 4 pCi/L was assumed for the radon concentration for this example. The results are shown in Figure 2.

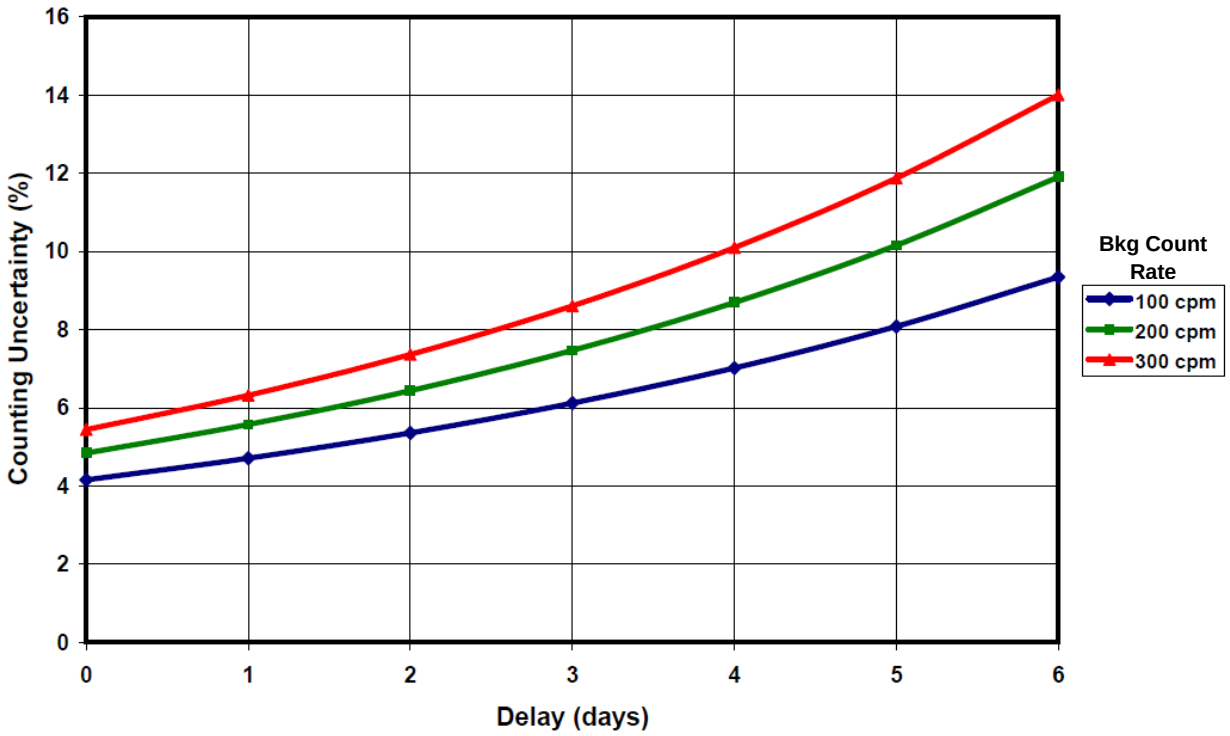

Figure 2. Effect of background count rate on counting uncertainty

After just one day of delay, S_{CT} ranges from 4.7% to 6.3% and increases rapidly after that. The counting uncertainty alone can equal or exceed 8% after about 2.5 to 5 days depending on the background count rate and with the assumed values in Table 1. S_{CT} can be a significant fraction of the total uncertainty, and with long delay times it can be the largest contributor to the total.

Effect of Relative Humidity

The values listed in Table 2 for several parameters were used to calculate the MDC for different values of calibration factor (CF) typical of a two-day exposure and for values of relative humidity of approximately 20%, 50% and 70%. The results are shown in Figure 3.

Table 2. Parameter values used for demonstrating effect of relative humidity on the MDC

Parameter	Value
Exposure time, t_e	48 hours
Counting efficiency, ε	0.39 cpm/pCi
Calibration factor, CF	0.12, 0.096 & 0.088 L/min
Sample counting time, t_s	10 min
Background counting time, t_b	10 min
Background count rate, cpm	200 cpm

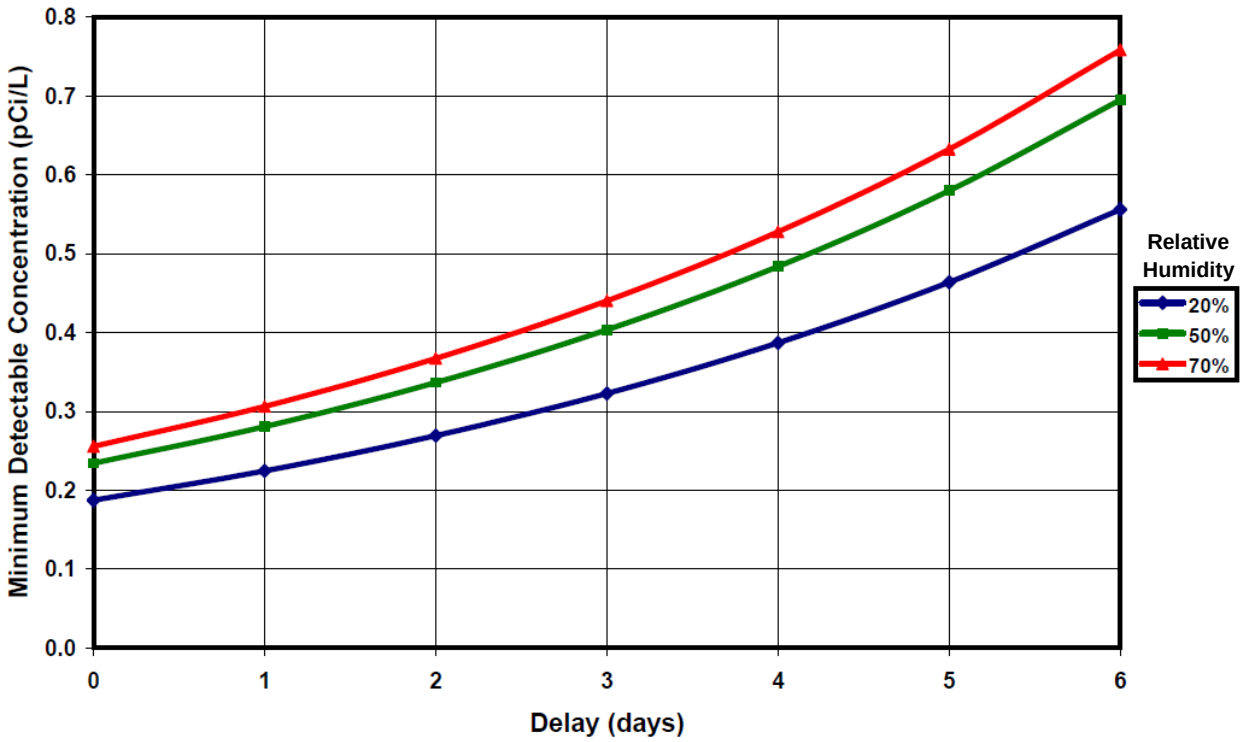

Figure 3. Effect of relative humidity on MDC

Again, the MDC increases with delay time. Note that the MDC increases with increasing relative humidity, because the value of CF decreases. The values of MDC were calculated using a background count rate of 200 cpm. If the background count rate were larger, then the MDC would be even greater.

The values in Table 2 were also used to calculate the counting uncertainty, S_{CT}. As was done above for the results shown in Figure 2, a value of 4 pCi/L was assumed for the radon concentration for these calculations. The results are shown in Figure 4.

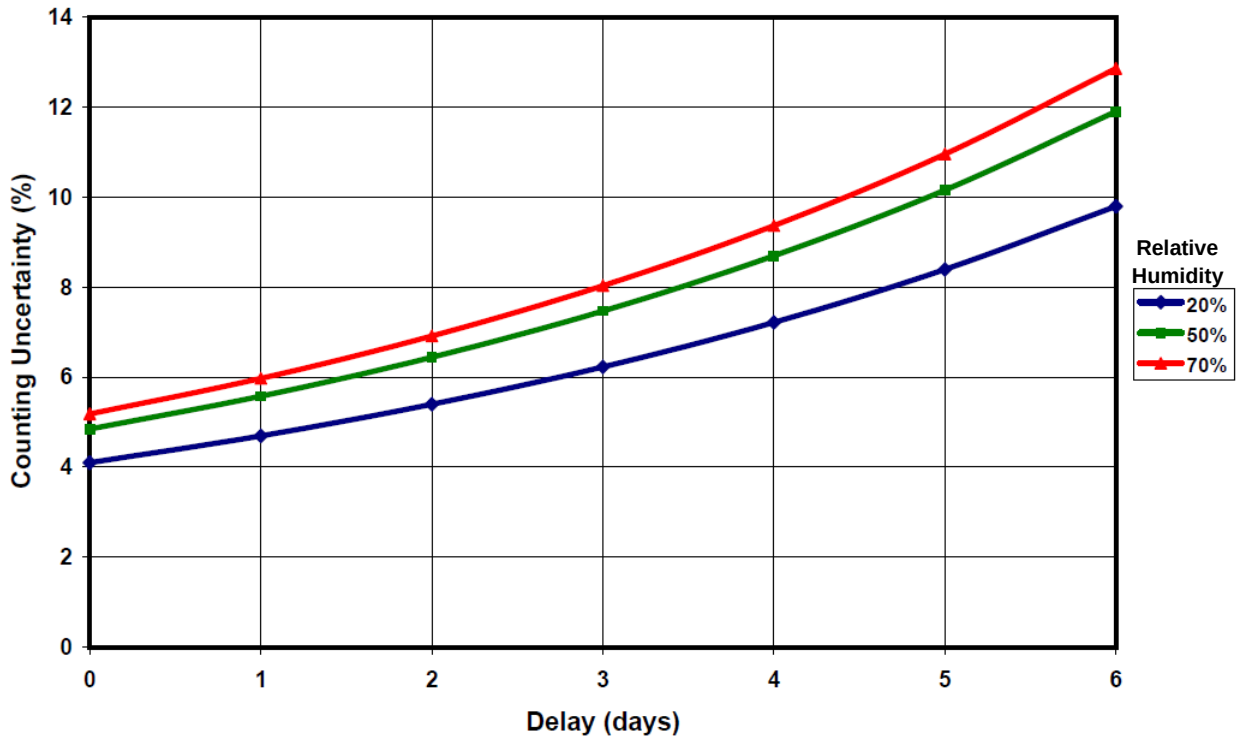

Figure 4. Effect of humidity on counting uncertainty

As was true for MDC, S_{CT} increases as the relative humidity increases, because less radon is adsorbed onto the charcoal at higher humidity.

Effect of Radon Concentration

The MDC is not a function of the radon concentration, but is a function of the "noise" or background. It is determined in the laboratory by counting a blank charcoal canister and depends on the specific analysis equipment and the times spent determining the background count rate and the sample gross count rate. However, S_{CT} is a function also of the "signal," which in this case is the gamma rays observed from the radon adsorbed on the charcoal and which in turn varies with the radon concentration. To demonstrate how S_{CT} changes with values of radon concentration, the parameter values in Table 1 were used to calculate S_{CT} for values of radon concentration of 2, 4, 6, 8 and 10 pCi/L. These results are shown in Figure 5.

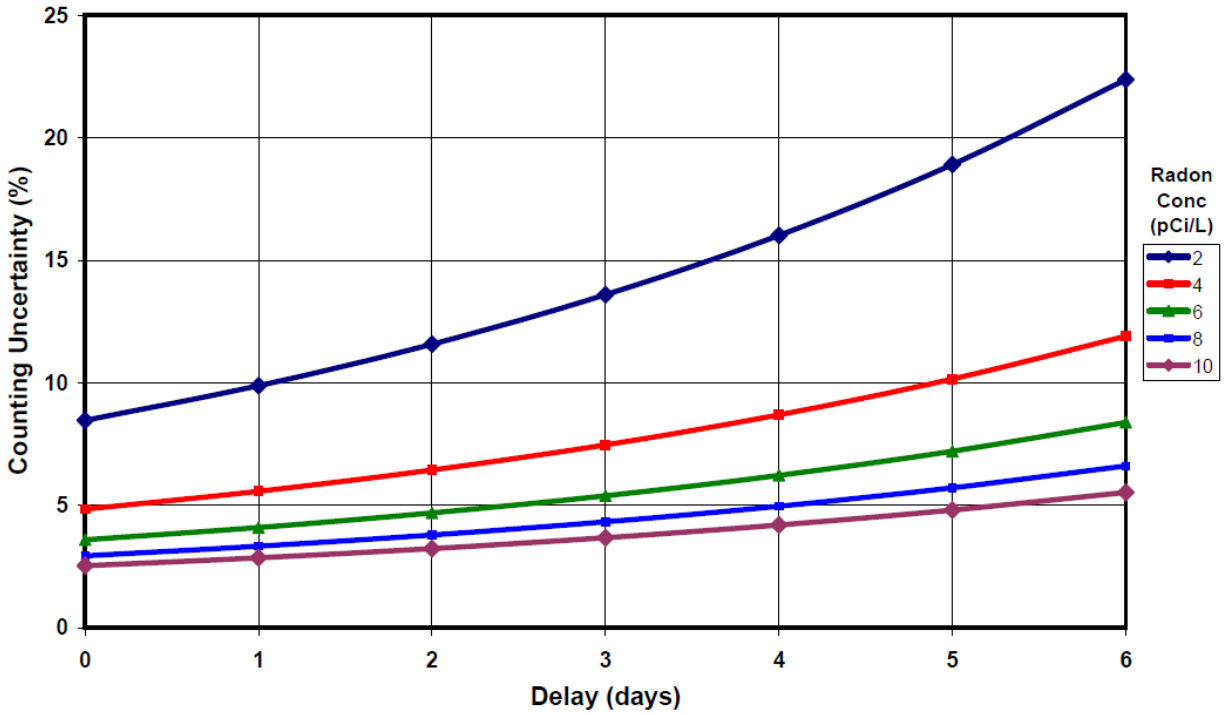

Figure 5. Effect of radon concentration on counting uncertainty

A background count rate of 200 cpm was used in the calculations; therefore, the curve corresponding to a relative humidity of approximately 50% in Figure 4 (the green curve) and the curve for 4 pCi/L in Figure 5 (the red curve) are identical. As expected, S_{CT} increases as the radon concentration decreases. Note from Figure 5, however, that below approximately 6 pCi/L, S_{CT} increases rapidly with decreasing radon concentration.

All analysis laboratories should have established control values for the MDC and S_{CT} for their measurements. For example, the laboratory may have a control value of MDC of 0.5 pCi/L and a value of 15% for S_{CT}. From the figures one can see that these values can be exceeded after just a few days of delay from the exposure period. Longer counting times would improve the signal-to-noise ratio, but a counting time of 10 minutes is typical. Combinations of factors such as high background and high humidity would make the situation even worse than shown in the figures.

Further, S_{CT} is only one component of the total uncertainty. The uncertainty associated with CF may be the largest contributor to the total uncertainty. As an example, assume that S_{CT} is 10% and that the uncertainty of CF is 15% and the uncertainties of all the other parameters are trivial. The total uncertainty from these causes alone would then be $(10\%^2 + 15\%^2)^{1/2}$ or 18%.

Some laboratories that analyze charcoal devices by gamma-ray spectroscopy use an equation that differs from equation (1) assumed here. However, similar results would be found regardless of the equation or model used; in other words, that MDC and S_{CT} both increase with delay time.

It is not possible to consider here all possible combinations of values of the various parameters for measurements using 4-inch open-face charcoal canisters or other charcoal devices. But a laboratory may apply the approach shown here to its measurements for charcoal devices that are

analyzed using gamma-ray spectroscopy. The underlying assumptions that make counting statistics valid are violated for several methods of measuring radon, but it has been shown (Jenkins, et al. 2006) that counting statistics may be applied to the analysis of charcoal devices using gamma-ray spectroscopy for the short counting times that are typically used. However, this is not true for analyses based on liquid scintillation spectroscopy. In order to calculate a valid S_{CT}, a correction must be applied to adjust counting statistics for these devices due to the detection of a significant number of correlated counts (Jenkins et al. 2006). Note that this reference contains a formula (equation 82) for calculating the counting uncertainty in cases where one or more of the assumptions that underlie counting statistics have been violated; however, there is an error in the equation as published. The following modification to equation (4) in this paper can be used for all charcoal devices or grab scintillation cells:

$$S_{CT} = 200 * \{[J R_s + (1 - J) R_b] / t_s + R_b/t_b\}^{1/2} / NCR \quad (6)$$

where J is a unitless term known as the "coefficient of dispersion." If J = 1, as it is for a Poisson distribution, then equation 6 reduces to equation 4.

Conclusion

The effects of background count rate and relative humidity on the MDC and S_{CT} with delay times ranging from 0 to 6 days were demonstrated through a few examples using typical values of several parameters for 4-inch open-face charcoal canisters. The effect of radon concentration on the values of S_{CT} was also demonstrated with the same range of delay time and typical values for parameters. The results demonstrated that both MDC and S_{CT} increase quickly with delay time, and can exceed acceptable levels in terms of control values or requirements of standards. Therefore it is important to analyze the sample as quickly as possible after the exposure, before the "signal" from the charcoal becomes indistinguishable from the "noise" of the background.

References

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