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TECHNICAL NOTE

A COMPARISON OF METHODS FOR PROMPT DETERMINATION OF POTENTIAL ALPHA ENERGY CONCENTRATIONS

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Abstract — The object of this study was to verify experimentally an optimisation procedure used to reduce the total time needed to obtain an accurate estimate of potential alpha energy concentration (PAEC) of ^{222}Rn progeny for a grab sampling method with only one count of gross alpha activity. Samples of air were analysed for potential alpha energy at concentrations near 2.08×10^{-6} J.m^{-3} (0.10 WL) using timing intervals of 7, 13, and 25 min. The measurements were compared graphically and statistically to estimates of PAEC obtained using the modified Tsivoglou method. No statistical differences in accuracy were found within or between the three total timing intervals. Suggestions for practical field instrumentation are presented.

INTRODUCTION

The concentration of radon gas in air is expressed in terms of radioactivity per unit volume. Normally the concentration of radon progeny is not expressed in terms of radioactivity per unit volume of air, but in terms of potential alpha energy concentration (PAEC). Potential alpha energy concentration is a measure of the total kinetic energy released by alpha particles from any mixture of radon progeny suspended in air that proceeds through the entire serial decay sequence down to ^{210}Pb. This concept is based on the premise that once the short-lived radon progeny are deposited in the respiratory system, they will remain there until the decay sequence is complete.

Historically, potential alpha energy concentration has been presented in units of working level. A working level (WL) is defined as

$$1 \text{ WL} = 2.08 \times 10^{-5} \text{ J.m}^{-3}$$

It is defined only for alpha emissions from the radon progeny and not from radon gas.

Prompt and accurate determinations of potential alpha energy concentration are desirable for health as well as economic reasons. Measurements in underground mines are required for purposes of estimation and control of exposure to radon progeny. Estimates of the annual exposure to miners are based on the combination of periodic grab samples and time of occupancy records[1]. Control of exposure refers to measurements used to confirm that a work area is below any required action level, especially following an incident such as an interruption of the ventilation system[2,3].

Regulations for the US mining industry state that persons shall not be exposed to air containing concentrations of radon progeny exceeding 2.08×10^{-5} J.m^{-3} (1.0 WL) or receive an annual exposure in excess of 0.014 J.h.m^{-3} (4 WLM)[4]. Weekly measurements are required in all areas of underground mines where the PAEC exceeds 6.24×10^{-6} J.m^{-3} (0.3 WL). The ICRP has made recommendations for an action level in the range of 500–1500 Bq.m^{-3} ^{222}Rn in all workplaces[5]. This corresponds to approximately 2.60×10^{-6} J.m^{-3} (0.125 WL). Thus, any measurement method used either for control or annual exposure assessment must be reliable in this range of concentrations.

The object of this study was to determine experimentally if the grab sampling procedures originally suggested by Borak[6] could indeed produce acceptable estimates of PAEC for total timing intervals less than 25 min at concentrations near the proposed action level of 2.08×10^{-6}J.m^{-3} (0.10 WL).

GRAB SAMPLING METHODS

A variety of grab sampling procedures have been developed to estimate PAEC. They are usually similar. Radon progeny suspended in air are collected on a membrane filter using a vacuum pump for a specified sampling interval (ST). The resulting activity on the filter is allowed to decay for a period of time (DT) and then counted during an interval (CT). The estimate of PAEC is based on the number of recorded counts. The total (TT) is the sum of the sample, decay and count times.

The most accurate estimate of potential alpha energy concentration would be obtained by integrating the total alpha energy released. This method is not practical because it would require up to 3 h for the short-lived radon progeny to decay entirely to ^{210}Pb.

The Tsivoglou method produces an estimate of potential alpha energy concentration, to which all other methods are normally compared[7]. The modified Tsivoglou method determines the concentrations of ^{218}Po, ^{214}Pb, and ^{214}Bi based on gross alpha counts in three separate counting intervals[8]. Potential alpha energy concentration can then be computed from the measured concentration of each of the radon progeny. The half-life of ^{214}Po is so short (164 μs) that it is assumed to be in radioactive equilibrium with ^{214}Bi.

Several procedures have been developed which derive PAEC from two independent measurements of activity collected on a membrane filter. An alpha spectroscopy method uses energy sensitive detectors to accumulate alpha counts in separate energy intervals from the decay of ^{218}Po (6 MeV) and ^{214}Po (7.7 MeV). Potential alpha energy is estimated as a linear combination of the two alpha counts that are multiplied by independent conversion factors[9,10].

Martz *et al*[11] proposed another alpha spectrometry method using two post-sampling times to measure count rates from ^{218}Po and ^{214}Po. This method yields estimates of PAEC based on four data points obtained over a total time of 35 min.

Alpha and beta particles from radon progeny can be measured simultaneously using two different detectors. The method consists of placing an alpha detector against the collection surface of the filter and a beta detector on the opposite side. The two gross count measurements are recombined in an equation to estimate PAEC where alpha and beta activity are weighted identically[12].

Holub[13] pointed out that this procedure did not take full advantage of the two independent pieces of available information. He modified this method by assigning a separate weighting factor for each count measurement in the estimation of PAEC. These methods have desirable features and are sufficiently accurate but require two counting systems. The beta detectors are sensitive to ambient photon exposure and thus must be properly shielded to maintain sensitivity below 2.08×10^{-5} $J.m^{-3}$ (1 WL).

Raabe and Wrenn[14] developed a method using maximum likelihood techniques for determining radon progeny concentrations in air. This method required many successive counts of gross alpha activity in order to establish the decay characteristics of the sample. The data are used in a fitting procedure to calculate each of the short lived progeny as well as PAEC. This method could produce estimates of potential alpha energy with greater accuracy than the Tsivoglou method with clearly defined estimates of uncertainties. The drawback was that it required counts to be taken over a period of time greater than one hour to characterise accurately the decay scheme of the progeny.

Kusnetz[15] developed a simple procedure to estimate potential alpha energy concentration from a single measurement of gross alpha activity collected on a membrane filter. The original Kusnetz method used a 5–10 min sample interval, during which instantaneous deposition of radioactivity on the filter was assumed. The sample was allowed to decay for 40–90 min and an alpha survey meter was used to determine the count rate after the decay interval. The estimate of potential alpha energy concentration was based on the measured count rate and decay time. The method proved to be acceptably accurate and used an inexpensive counting system that was insensitive to fluctuating gamma ray background. However, it required a long decay time and thus a long total time interval.

The Kusnetz method has been modified by Breslin[16] to eliminate a rate meter. Rolle[17] suggested procedures based on mine tunnel models for reducing measurement times. He was able to identify timing intervals where the estimate of PAEC could be obtained within 20 min or less.

Borak *et al*[18] generalised the single gross alpha method by not assuming instantaneous deposition of activity on the filter not requiring instantaneous determination of count rate. He also presented an optimisation procedure that included variations in the mixtures of progeny as well as the uncertainty due to counting statistics[6].

EXPERIMENTAL METHODOLOGY

A series of measurements were performed at the Bureau of Mines, Denver Research Center. Grab samples were taken from a radon test chamber with a volume of 3.9 m^3[19]. Radon progeny concentrations were controlled by adjusting the concentration of radon gas together with condensation nuclei. A continuous beta detection monitoring system was utilised to estimate the potential alpha energy concentration in the test chamber[18]. In addition, estimates of PAEC and radon progeny mixtures were determined periodically with the Tsivoglou method.

The chamber was equipped with several ports through which filter holders could be placed for obtaining grab samples. Air was passed through a membrane filter 2.54 cm in diameter with a pore size of 0.8 μm. Air flow was maintained by a metal bellows pump and the total volume of flow was measured with a dry test meter. The sample filters were removed and placed with the collection surface down directly on a Mylar film coated with zinc sulphide. The film was positioned on the face of a 7.6 cm diameter photomultiplier tube (PMT). The signals from the PMT were amplified and recorded by a multichannel analyser operating in the multiscale mode. The system response was assumed to be independent of alpha energies ranging from 4 to 8 MeV.

A background count was taken periodically to estimate the build-up of gross alpha activity on the Mylar film. A new film had a background count rate of less than 1 count per minute. The film was replaced if the background count rate exceeded 15 counts per minute.

PAEC was determined using the following formula:

$$\text{PAEC } (\text{J.m}^{-3}) = \frac{C_\alpha}{\epsilon \times \text{VPM} \times \text{ST} \times \text{CT} \times \text{K}} \quad (1)$$

where

C_α = net alpha counts,
ϵ = absolute counting efficiency,
VPM = flow rate (l.min^{-1}),
ST = sampling interval (min),
CT = counting interval (min),
K = conversion factor ($\text{dpm}\cdot\text{m}^3\cdot\text{l}^{-1}\cdot\text{J}^{-1}$).

The absolute counting efficiency, ϵ, of the system was determined daily using a ^{230}Th check source (4.7 MeV) traceable to the NIST. The flow rate of air, VPM, that flowed through the filter per unit time was measured for every grab sample.

The conversion factor, K, for each timing sequence was determined with a computer program that calculates alpha disintegrations from a mixture of radon progeny that is collected on a filter. This specific mixture is one typically found in uranium mine atmospheres and is equivalent to 8290, 3770 and 2480 Bq.m^{-3}; for ^{218}Po, ^{214}Pb and ^{214}Bi respectively, at a PAEC of 2.08×10^{-5} J.m^{-3} (1 WL)[19]. The conversion factors are then determined by rearranging Equation 1.

Measurements of PAEC were obtained for three total time intervals, TT, and several different decay intervals, DT, for each TT (Table 1). It was not considered practical to obtain estimates of PAEC for decay times less than 1 min, because the filter had to be transferred manually from the filter holder to the counting system. In all cases, ST = CT, because the largest number of alpha disintegrations, thus greatest number of counts, occurs when the sampling and counting intervals are equal for a fixed decay interval[18,21].

Table 1. Timing intervals (min) used to estimate PAEC.

Total time (TT)	Sample time (ST)	Decay time (DT)	Count time (CT)
7	3.0	1.0	3.0
	2.5	2.0	2.5
	2.0	3.0	2.0
	1.5	4.0	1.5
	1.0	5.0	1.0
13	6.0	1.0	6.0
	5.0	3.0	5.0
	4.0	5.0	4.0
	3.0	7.0	3.0
25	12.0	1.0	12.0
	11.0	3.0	11.0
	10.0	5.0	10.0
	9.0	7.0	9.0

Five grab samples were taken at each timing interval. The arithmetic mean of the five estimates of PAEC was compared to the Tsivoglou measurement that was taken at the time closest to the group of grab sample measurements.

The following method was used to identify systematic biases in the grab sample procedures. The mixture of progeny in the chamber as determined by the Tsivoglou method was processed through the computer program described above to determine the expected net alpha counts for a given timing sequence. The result was used in Equation 1 to obtain a predicted estimate of PAEC for this mixture and timing sequence. This predicted value of PAEC reflects the inherent error associated with using the standard value of K defined for the mixture typically found in a mine.

RESULTS AND DISCUSSION

Grab sample measurements were taken over nine days. The concentration of condensation nuclei ranged from 1.25×10^5 to 2.40×10^5 (particles.cm^{-3}) and varied by no more than 3% over a measuring period. The relative humidity in the test chamber varied by no more than 2%. The air flow rate through the membrane filter was 2.3 ± 0.1 l.min^{-1}. The counting efficiency for the ZnS scintillator using the ^{230}Th check source was $48 \pm 1\%$.

The mean pressure difference across the filter was 31 ± 3 kPa. The pressure difference indicated how tightly the filter fitted against the filter holder and the existence of holes in the membrane filter. Both of these problems would cause an overestimate of the volume of air that flowed through the filter, resulting in an underestimate of potential alpha energy concentration[2]. The filter was replaced if the pressure fell below 24 kPa.

The radon progeny concentrations that were determined from the Tsivoglou method over the course of the experiment were normalised to 2.08×10^{-5} J.m^{-3} (1 WL) and an arithmetic mean for each daughter was calculated. The mean concentrations were: 8770 ± 1700, 3770 ± 260, 2370 ± 480 Bq.m^{-3} for ^{218}Po, ^{214}Pb, and ^{214}Bi, respectively. This was not significantly different from the typical mixture found in underground mines[20].

A total of 227 grab sample measurements were taken. Data were collected at $\sim 2.08 \times 10^{-6}$ J.m^{-3} (~0.1 WL) for total times of 7, 13, and 25 min; at $\sim 9.4 \times 10^{-6}$ J.m^{-3} (~0.45 WL) for total times of 13 and 25 min; and at $\sim 1.9 \times 10^{-5}$ J.m^{-3} (~0.9 WL) for a total time of 7 min. Figures 1, 2, and 3 show the estimates of PAEC for the three total time intervals and associated decay intervals plotted against the values obtained with the Tsivoglou method. The error bars represent one standard deviation of the sample of five measurements taken for a given timing interval.

This graphical comparison indicated that there were no apparent differences in accuracy between the prompt grab samples and Tsivoglou measurements for total times of 7 and 13 min. However, Figure 1 shows a slight tendency to underestimate PAEC for the data near 10^{-5} J.m^{-3} at TT = 25 min.

Figure 4 shows values for the prompt measurements plotted against the predicted estimates of PAEC. There were no large differences between these values. The measurements that had apparently underestimated potential alpha energy concentration in Figure 1 were comparable with their predicted values. Therefore, the prompt estimates of PAEC taken at TT = 25 min would typically be inaccurate for the progeny mixture that was present in the test chamber at that time. These results imply that there were no systematic errors in the sampling procedures.

An analysis of variance (ANOVA) was used to determine if there were statistical differences in the accuracy of the prompt estimates of PAEC for the different concentrations and various timing intervals. The accuracy of each prompt measurement of PAEC was defined as follows:

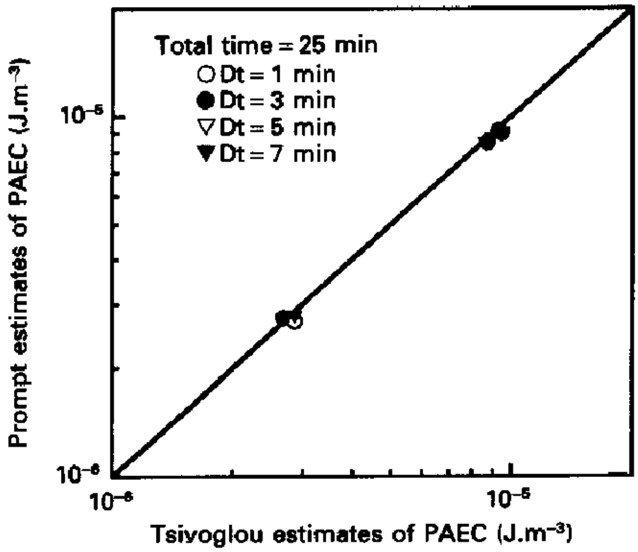

Figure 1. Comparison of prompt estimates of PAEC for total timing intervals of 25 min with values obtained using the Tsivoglou method.

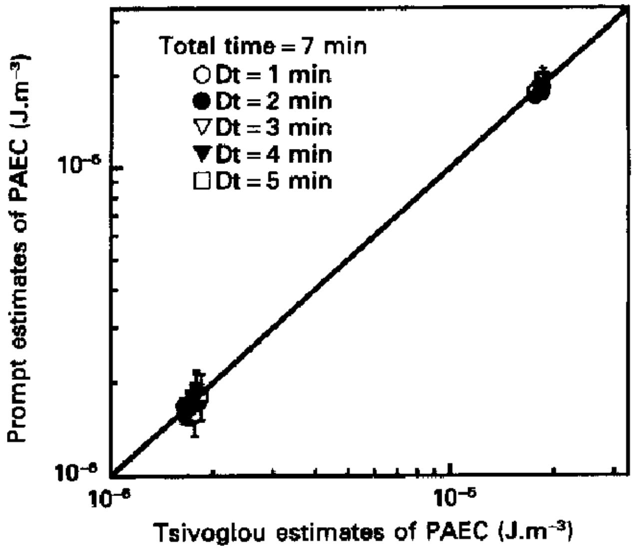

Figure 3. Comparison of prompt estimates of PAEC for total timing intervals of 7 min with values obtained using the Tsivoglou method.

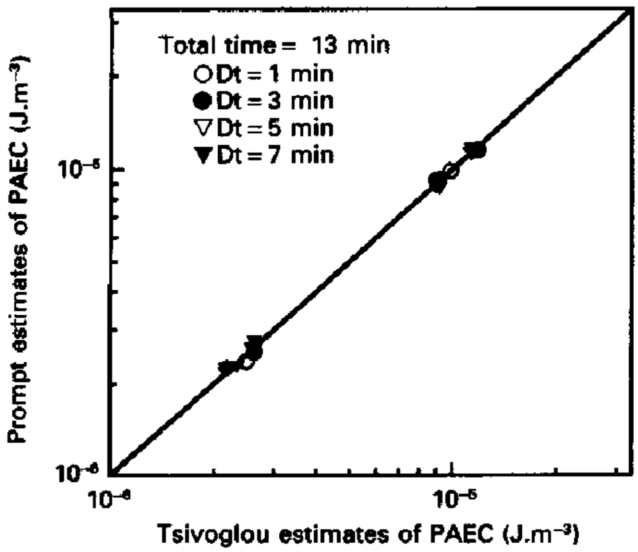

Figure 2. Comparison of prompt estimates of PAEC for total timing intervals of 13 min with values obtained using the Tsivoglou method.

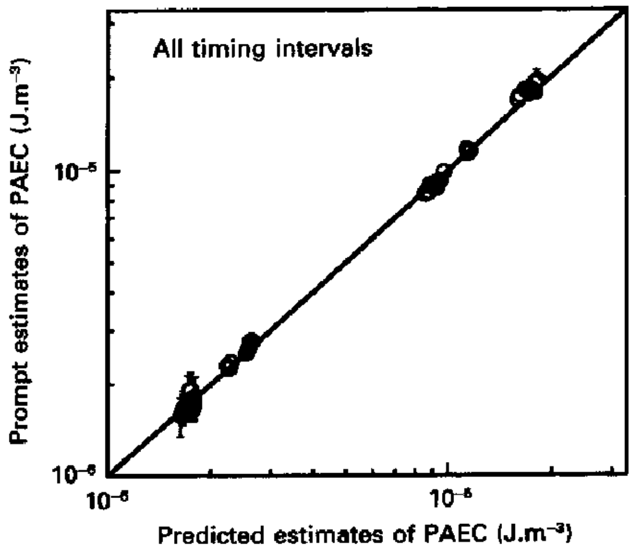

Figure 4. Comparison of the prompt estimates of PAEC for all total timing intervals with the predicted values for the corresponding mixture of ^{222}Rn progeny in the test chamber.

$$\Delta_i = \frac{\text{Prompt estimate}_i - \text{Tsivoglou estimate}_i}{\text{Tsivoglou estimate}_i}$$

This measure of accuracy was modelled as a linear combination of PAEC, decay time, and total time to obtain a predicted value of accuracy:

$$\tilde{\Delta}_i = K_0 + K_i PAEC_i + K_2\, TT_i + K_3\, DT_i$$

An ANOVA Type III sum of squares for an unbalanced model design was used to determine if the accuracy of the prompt measurements, Δ_i, could be predicted by any of the variables in Equation 3[22]. None of these parameters was significant at the 95% level of confidence, $p < 0.05$.

This type of ANOVA requires that the residuals ($\Delta_i - \tilde{\Delta}_i$), be normally distributed with constant variance. A scatter plot of ($\Delta_i - \tilde{\Delta}_i$) against $\tilde{\Delta}_i$ indicated that these conditions may not be satisfied. A square root transformation of Δ_i stabilised the variance. The ANOVA was repeated with the transformed data and yielded the same results.

A least-squares means function (multiple t-tests on means for unbalanced design) was performed on the model parameters to determine if there were any interactions between the parameters themselves[22]. There were no signifcant interactions at $p < 0.05$.

CONCLUSION

The object of the study was to evaluate the accuracy and precision of prompt estimates of PAEC for total time intervals of 7, 13, and 25 min. A generalised gross alpha method was applied to grab samples of air taken at three values of PAEC for these total timing intervals and several different decay intervals. Direct comparisons were made between the prompt estimates of PAEC and values obtained from the Tsivoglou method.

The results showed that the total time could be reduced to 7 min without a significant loss in accuracy for a typical mixture of radon progeny found in uranium mine atmospheres at PAEC levels that ranged from $\sim 2.08 \times 10^{-6}$ J.m^{-3} (0.1 WL) to $\sim 1.9 \times 10^{-5}$ J.m^{-3} (0.9 WL). An analysis of variance did not identify any differences as a function of decay time, DT.

The method used to analyse these experimental data was not identical to the optimisation process used by Borak[6] because it was not possible to generate 250 random combinations of radon progeny in the test chamber. However the data did confirm that prompt measurements can be made for typical mixtures near the proposed action levels. The additional uncertainty resulting from variations in the mixture of radon daughters in underground atmospheres is ~5% at TT = 25, ~6% at TT = 13 and ~12% at TT = 7[6].

Borak[6] showed that a decay time of 3 min would optimise the combined uncertainties associated with random mixtures and counting statistics. Although no effect was observed as a function of decay time, it is suggested that 3 min is an appropriate value both for minimising uncertainties and convenience for transferring the filter to the counting system.

These results imply that a simple method can be used to measure PAEC in underground mines with an acceptable level of uncertainty for compliance with existing and proposed regulations. The grab samples can be collected with a portable pump producing a flow rate of ~ 2 l.min^{-1} that can meet safety requirements for underground mines. The sampling system can be designed with an open-faced filter to eliminate plateout. An operator might manually transfer the filter from the sampling head to the counting system, or everything could be combined into one unit.

A gross alpha counting system has the advantage that it is not sensitive to interference from photon exposure rates underground. Various detectors could be used, but the combination of counting efficiency and flow rate should be selected such that: $\epsilon\, VPM \approx 1$, to yield the same precision (i.e. counting statistics) obtained here and recommended by Borak[6]. A small microprocessor could control the timing intervals and process the data immediately. Appropriate safety precautions would have to be included for intrinsic spark prevention if used in underground coal mines.

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