

Chapter 7. Radon and Radon Daughter Monitoring
(including thoron daughter monitoring)

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Abstract. Radon/radon daughter and thoron daughter measurement techniques are outlined. The necessary precautions and critical assessments of each method are also presented with a view to providing a better understanding of the various measurement methods.

INTRODUCTION

The radiation monitoring programme in mines where radioactive ores are extracted requires the measurement of radon daughters and thoron daughters as these account for much of the radiation exposure of employees. A monitoring programme has two objectives. Firstly it provides quantitative information which allows for satisfactory engineering control of radon or thoron daughter concentrations in mine air, and secondly there is a legal requirement for operators to maintain complete records of cumulative exposure for all employees. This relates of course to the operator's obligation to ensure that employee exposures do not exceed the limits laid down under the Code of Practice (Australian Government Publishing Service, 1980). Such records are also likely to be used for future epidemiological studies of the effects on occupational health of radon and thoron daughter exposures.

At the present time, cumulative exposure to radon or thoron daughters is assessed in units of Working Level Month (see Chapter 1 for a definition of WLM) and is obtained by maintaining adequate records of working levels at the various work stations, and combining these with individual employee records which define the period spent by the employee at each work station. However this method is only adequate if area measurements are made frequently enough to follow major fluctuations in daughter levels. In time it is likely that this method will be replaced by personal dosimeters, worn by each employee, and which will provide a measure of integrated exposure just as the TLD dosimeter provides an assessment of integrated exposure to external radiations. With well co-ordinated record keeping, it is reasonable to expect that a radiation safety officer should be able to provide integrated radon or thoron daughter exposures in terms of Working Level Months, with an overall uncertainty of less than 50 per cent.

Some General Considerations

As is the case for most measurement practices, good housekeeping and a thorough understanding of the science behind the methods employed are essential if consistent and reliable results are to be obtained. A separate clean laboratory is desirable both for the calibration and maintenance of field instruments, and for the analysis of field samples. Calibration facilities for checking flow rates in air sampling equipment for example are just as important as facilities for determining the energy response and efficiency of radiation survey instruments.

Ideally, radiation protection staff will have a sound practical understanding of working conditions throughout the mine or mill, of usage and effectiveness of control equipment (e.g. ventilation equipment) and will be aware of any other operational factors likely to have a bearing on their interpretation of measurements or on the best management of radiation protection. In some cases, radiation measurements - for example estimates of radon emanation rates from freshly exposed ore - can be helpful and may assist the mine engineers in their planning of methods, required ventilation and so on, for the optimum management of radiation related parameters.

PHYSICAL CONSIDERATIONS

Radon-222 and its decay chain

The decay chain of which ^{222}Rn is a part is one of the oldest and best-studied phenomena in nuclear physics. It begins with the naturally occurring isotope ^{238}U and ends with the stable ^{206}Pb . In between there are 8 alpha decays and 6 beta decays. More detailed data are given in Table 1. Note that many of the chain members are still often referred to by their historical names.

Radon-222 provides a natural division for our purposes, because we are interested mainly in radon and its daughters. However, there is also an important physical basis for this separation - namely the fact that radon is an inert gas. As ^{238}U decays through five steps to ^{226}Ra , the heavy nucleus remains fixed in the rock. However, there is diffusion of radon gas from the local site of production, and in any particular (surface) rock some of the radon will escape into the surrounding atmosphere.

Thus, for many practical purposes one can think of uranium laden ore as a source of radon gas, (although in fact ^{226}Ra is the true source) as well as a source of the many α , β and γ radiations being emitted by the constituent nuclei themselves.

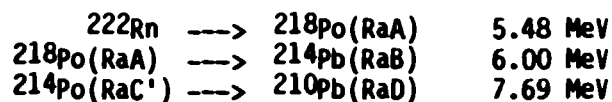
Let us examine the properties of the chain from radium to radon-daughters. We note that ^{226}Ra has a half-life of 1602 yr. Since this is much longer than the half-life of any subsequent daughter, ^{226}Ra can be viewed as a fixed-rate generator for ^{222}Rn , whose half-life is 3.83 days. Radioactive equilibrium between the succeeding daughters (RaA, RaB, RaC) is achieved after approximately three hours. Note that after RaC decays, RaC' follows nearly immediately (164 μsec) while RaD (^{210}Pb) has a 22-yr half-life, which for short times effectively blocks the decay chain. Thus, we can deal here exclusively with the first four daughters, RaA, RaB, RaC and RaC'.

Evans (1969) has given a very useful summary of the physical and engineering considerations relevant to the control of the radon daughters. Although radon is inert, the three elements immediately below it in the periodic table (polonium, bismuth and lead) are all chemically active. In particular, when radon (gas) decays, most of the newly created ^{218}Po (RaA) atoms, which are typically ionized, tend to attach almost immediately to any particulate matter in the atmosphere. Most RaB, RaC and RaC' atoms are usually attached as soon as they are formed. In dust-laden mine air, the consequence is that the particulate matter becomes radioactive by adsorption. Whether or not they are attached to particulate matter, the daughters cause important radiological consequences when inhaled, since they tend to lodge in the lung mucosa.

TABLE 1. Main sequence of decays from ^{238}U to ^{206}Pb .
Three very weak collateral branch disintegrations,
all with branching ratios less than 0.1%, are omitted.

Common name or symbol	Half Life	Principal radiations	Alpha energy (MeV)	Gamma-ray quanta per decay	Average Gamma-ray energy (MeV)
Uranium I - ^{238}U	4.49×10^9 yr	α	4.18		
Uranium X ₁ - ^{234}Th	24.1 days	β			
Uranium X ₂ - ^{234}Pa	1.17 min	β			
Uranium II - ^{234}U	248,000 yr	α	4.76		
Ionium - ^{230}Th	80,000 yr	α	4.68 (75%) 4.61 (25%)		
Radium - ^{226}Ra	1,602 yr	α	4.78 (94.3%) 4.69 (5.7%)		
Radon - ^{222}Rn	3.825 days	α	5.485		
Radium A - ^{218}Po	3.05 min	α	5.998		
Radium B - ^{214}Pb	26.8 min	β			
		γ		0.82	0.295
Radium C - ^{214}Bi	19.7 min	β			
		γ		1.45	1.050
Radium C' - ^{214}Po	164 μsec	α	7.69	1.0	0.047
Radium D - ^{210}Pb	22 yr	β			
		γ			
Radium E - ^{210}Bi	5.02 days	β			
Radium F - ^{210}Po	138.3 days	α	5.298		
Radium G - ^{206}Pb	Stable	Stable			

The alpha-emitting daughters are RaA and RaC'. The alpha decay energies are as follows:



The energy difference can, of course, provide a basis for distinguishing the individual components by alpha spectroscopy.

The two important beta/gamma-emitting daughters are RaB and RaC. Each has a rather complicated set of emissions, with three important betas and several gamma lines each (see Table 2). Because of this, beta detection as a means of measurement is complicated as the efficiency of a beta detector is very hard to determine. The RaB and RaC gamma lines can be detected by gamma-spectroscopic methods but this is not now commonly done.

TABLE 2. Radon decay series

Isotope	Half-life	Principal decay modes
${}^{222}\text{Rn}$ (radon)	3.824 d	α (5.49 MeV, 100%)
${}^{218}\text{Po}$	3.05 m	α (6.00 MeV, 100%)
${}^{214}\text{Pb}$	26.8 m	β (1.04 MeV, 7%; 0.75 MeV, 43%; 0.69 MeV, 46%; others)
		γ many
${}^{214}\text{Bi}$	19.8 m	β (3.28 MeV, 19%; 1.90 MeV, 8.5%; 1.43 MeV, 8.7%; many others)
		γ many
${}^{214}\text{Po}$	162 μs	α (7.69 MeV, 100%)

Radon-220 (thoron) and ${}^{219}\text{Rn}$ (actinon)

In situations where ${}^{222}\text{Rn}$ must be measured, two other isotopes of radon may interfere with the measurement. The most important is radon-220 (historically known as 'thoron'), a member of the decay chain which originates with naturally-occurring ${}^{232}\text{Th}$. The other isotope is radon-219 (historically, 'actinon'), from the chain originating with ${}^{235}\text{U}$. Both of these gases behave in a similar manner to ${}^{222}\text{Rn}$ and emanate from the radioactive ores after their birth following alpha decays of their immediate parents (${}^{224}\text{Ra} \longrightarrow {}^{220}\text{Rn}$, ${}^{223}\text{Ra} \longrightarrow {}^{219}\text{Rn}$).

In Australian uranium mines, the thorium content of ores is generally low (typically 10% of that of ${}^{238}\text{U}$), and the ${}^{235}\text{U}/{}^{238}\text{U}$ ratio is uniform at about 0.71% (CRC Handbook 1979-80). Thus the equilibrium production of both thoron and actinon gases is relatively small. Equally important the half-lives are quite short (55 s. for thoron, 4.0 s. for actinon). Because of their limited abundances and short half-lives, their direct radiological impacts are in both cases much less significant than that of ${}^{222}\text{Rn}$.

The thoron daughters ^{212}Pb and ^{212}Bi have half-lives of 10.6 and 1.01 hr, respectively and extraneous-counts can be recorded when ^{222}Rn daughters are collected on air filters for measurement. Because of actinon's short half-life, its ability to diffuse out of earth and rock is so limited that it is seldom, if ever, present at levels measurable in the environment. The thoron decay series is shown in table 3.

TABLE 3. Thoron decay series

Isotope	Half-life	Principal decay modes
^{220}Rn (thoron)	55.3 s	α (6.28 MeV, 100%)
^{216}Po	0.15 s	α (6.78 MeV, 100%)
^{212}Pb	10.64 h	β (0.57 MeV, 19%; 0.33 MeV, 77%; 0.15 MeV, 49%) γ many
^{212}Bi	60.6 m	α (5.61-6.07 MeV, 33.7%) β (2.25 MeV, 54.8%; 1.52 MeV, 5.0%; others) γ many
^{212}Po	0.3 μs	α (8.78 MeV, 66.3%)
^{208}Tl	3.07 m	β (1.80 MeV, 18.7%; 1.52 MeV, 8.2%; 1.29 MeV, 8.5%; 1.03 MeV, 1.0%) γ many

METHODS OF MEASURING RADON EMANATION FROM URANIUM ORE

The Accumulator Method

This method was first introduced by Thompkins and Cheng (1969) who placed a large drum (44 gallon) over the area under investigation. The drum is usually grouted to a flat surface to prevent radon loss at the collection interface. Radon gas diffuses from the ore into the drum. A steady state is reached after approximately 3 hours by which time the radon concentration in the drum equals the radon concentration in the soil pore air space. The radon emanation rate will decline as the partial pressure of radon in the drum approaches that in the pore air space. Thus, the radon in the drum should be collected as soon as the drum is placed over the measurement area. A flushing mechanism can be used to remove radon from the drum prior to further measurements.

The radon concentration in the drum can be determined by using the two-filter tube method or by taking air samples using an evacuated Lucas cell (Lucas 1953) as described below. If Lucas cells are used to sample the air in the drum an internal fan must be fitted to ensure that the heavy inert radon in the drum is mixed prior to sampling. The in-situ radon emanation rate is simply the radon flux per unit area and is calculated by the following formula:

$$E = \frac{CV}{\Delta TA}$$

where E is the radon emanation rate ($\text{Bq m}^{-2}\text{s}^{-1}$)
 C is the radon concentration in the drum (Bqm^{-3})
 V is the volume of the drum (m^3)
 ΔT is the time elapsed during accumulation of radon in drum (s)
 A is the area of the drum (m^2)

The Charcoal Method

This method utilizes the adsorption capacity of activated coconut charcoal. Cylindrical canisters, typically 6cm in diameter and 6cm high are filled with freshly degassed granulated charcoal which is held in position with a wire mesh, supported by a circlip. To ensure that previously adsorbed moisture and other gaseous products are removed prior to use, the charcoal is degassed by placing the canister on top of a hot plate ($300\text{--}400^\circ\text{C}$). When the canisters have been degassed and cooled to ambient temperature they are placed in a plastic bag and sealed before use.

In the field, a number of canisters are placed over the area under investigation. Canisters are placed or embedded into soft ground by twisting to a depth of 1cm with additional soil packed around the edge to prevent leakage. Often in hard rock the canister must be fixed onto the surface.

The radon collection time is determined by the emanation rate and the dampness of the area. Charcoal adsorbs water vapour and this reduces its capacity to adsorb radon. However, Countess (1977) found that when charcoal adsorbs 110% by weight of water its capacity to adsorb radon is reduced by less than 10%. If the rock has a high moisture content, the charcoal efficiency for collecting radon could be reduced when using long collection times. Providing the charcoal is not saturated and is still on the linear part of the adsorption curve (Little, 1975) then the collection efficiency is unaltered. Thus, canisters placed at the same location for short and long periods should yield the same result for the calculated emanation rate. If the charcoal becomes saturated then a substantial drop in the calculated radon emanation rate will be observed at the longer exposure times.

The measurement of the canisters is usually performed with a 5cm x 5cm Sodium Iodide (NaI) crystal connected to a single channel or multichannel analyser to detect the 609 keV gamma ray of ^{214}Bi (RaC). The 609 keV gamma ray is chosen because of its excellent separation from other overlapping emission lines, and the generally low detector background in this energy region. For greater sensitivity, counting is often performed 3 hours after collection to allow the radon daughter products to come into equilibrium with the parent radon prior to counting.

The canisters must always be counted in the same geometry in order to ensure reproducibility of results. The calibration procedure is as follows: An exposed canister is counted and then its charcoal is degassed into a radon analyzer (Lucas 1964). The radon collected in the analyzer is then transferred to a Lucas cell for counting. Typically, the counting efficiency of the canister with a NaI (5cm x 5cm) detector is about 1.5% for the 609 Kev gamma ray.

This method does not suffer from the equilibrium problems of the accumulator method since it does not distort the radon partial pressure immediately above the surface. This is because the radon, being adsorbed on the charcoal, is removed from the air space below the charcoal. Countess (1977) seals the canisters during storage prior to counting because he has found that adsorbed radon is lost at the rate of 0.25% per hour at 23°C .

This rate of loss increases with temperature and decreases with increasing pressure. Our experience has been that the activity loss at room temperature (22°C) is approximately 20/o per hour.

The radon emanation rate can be calculated from the following equation:

$$E = \frac{X \lambda^2 e^{\lambda t_e}}{cA (1 - e^{-\lambda t_e}) (1 - e^{-\lambda t_c})}$$

where E is the radon emanation rate (Bqm⁻²s⁻¹)
 X is the counts registered above background
 λ is the radon decay constant (2.1 x 10⁻⁶s⁻¹)
 ε is the efficiency of detection (counts/disintegration)
 A is the area of the canister (m²)
 t_e is the elapsed time between the end of exposure and the start of counting
 t_c is the exposure time of the canister
 t is the counting time

Emanometers

These instruments use a solid state alpha detector at the top of a canister which is placed on the ground. The detector is connected to counting electronics and has a digital readout to give integrated counts.

Radon diffuses into the canister and unattached Radium-A (²¹⁸Po) plates out (Kaufman and Dinwiddie 1971) on the surface of the detector and emits an alpha particle which is registered as a count. The number of counts registered is related to the radon concentration in the canister.

This instrument has been traditionally used for exploration rather than absolute emanation rate measurements. However, we have no experience of the reliability or stability of calibration of this type of instrument, if it is to be used for the latter purpose.

MEASUREMENTS OF RADON CONCENTRATIONS IN AIR

Scintillation Cells

The simple ZnS (Ag) coated cell developed by Lucas (1953) has been very useful for measuring the radon concentration in air.

Scintillation cells can be made of various sizes from 100 ml to 1 litre. The design we prefer, and which is described in detail in Leach and Lokan (1979), consists of a copper cell with a glass end window. For very low background applications the end window on the scintillation cell should be made of quartz glass. However, in practice, radon concentrations in air in uranium mines are high so that inexpensive plate glass is adequate. The inside walls of the metal housing are coated with ZnS (Ag) silver activated powder. The coating of ZnS on the walls of the cell is applied by dusting the powder on to a thin coating of clear lacquer. Excess powder is removed from the cell.

Depending on the cell efficiency required, the end windows may be treated in two ways before being glued to the metal housing. Lucas (1953) coated the front glass windows with a transparent electrically conductive coating of tin oxide to reduce the effects of stray electric fields which can cause selective plate-out of radon daughters on the walls of the cell. A typical efficiency of a 100 ml cell of the above type is 750/o (5 cpm/pCi).

The application of a conducting layer to the glass requires special facilities. However, a simple alternative is to coat all inside surfaces with scintillator. This leads to a reduction in efficiency of approximately 20%, but this is of little consequence in most practical situations.

The cells are thoroughly evacuated prior to use and the sampling of mine air consists of opening a cell to the atmosphere and resealing it. When the cells contain radon, radon daughters plate out onto the ZnS (Ag) coating on the inside wall of the metal housing. Although the cell is evacuated after counting, the background is still high due to plate out of daughters and usually a period of three hours is needed before the cell may be reused to allow existing daughter products to decay. Depending on the frequency of use and activity of radon sampled in each cell, the background of the cell will increase due to build-up of ^{210}Po (Ra F). Thus, cells have to be recoated after prolonged use.

The efficiency may vary slightly from cell to cell but this variation does not normally exceed $\pm 10\%$. The calibration of individual cells requires access to a standard radium-226 solution from which radon can be drawn. This can be carried out satisfactorily only under well controlled laboratory conditions (Lucas 1964).

After sampling the air with an evacuated cell, the alpha scintillations are detected by placing the end window of the cell directly on top of a photomultiplier tube (PMT) in a light tight enclosure. Pulses from the PMT are amplified and counted with a discriminator/scaler.

The scintillation cell design has been modified for use as a continuous sampling device. Filtered air is drawn through the cell by a pump and the number of alpha scintillations observed by a photomultiplier are recorded in a preset counting interval. Air sampling rates of approximately 30 l/min. are used, causing turbulent air movement through the cell, which enhances plate-out of the unattached RaA atoms. The efficiency of such cells is usually about 60% (4cpm/pCi) with a lower limit of detection of around 0.04 Bq/l (1 pCi/l). The main disadvantage of these cells is that the short-lived RaA decays through to RaC which has a half-life of 19.7 minutes. Thus, this monitor cannot resolve rapidly changing radon concentrations but smoothes out changes with about a 30 minute time constant.

Although reliability is an important attribute of the scintillation cell, several precautions are necessary to avoid errors. In this regard the following should be pointed out:

- (a) plastic chambers are susceptible to loss of radon by diffusion through the cell walls because radon diffuses readily, if slowly, through plastic materials. However, the rate of loss over a period of a few hours is generally negligible.
- (b) imperfect valves are another cause of sample loss. If a good vacuum is not maintained less than the full cell volume is used for collection.
- (c) deposition of radon daughters on non-conductive chamber walls (glass, plastic) may be uneven and result in variation in counting efficiency, although this effect is remedied by the application of a conductive coating.

Two-filter Tube

The two-filter method was developed by Thomas and Le Clare (1970). This method uses a cylindrical tube which is fitted at both ends with filter holders for collection of radon daughters from the air so that only radon enters the tube. As radon progresses down the inside of the tube, RaA is produced and a fraction is collected at the exit filter. The radon daughter activity on the inside of the exit filter is proportional to the radon concentration in air. This method provides a rapid way of determining radon concentrations in air, with a lower limit of detection of approximately 0.4 Bq/l (10 pCi/l).

Large two-filter tubes with much higher sensitivities have been constructed so that environmental background radon and thoron concentrations can be measured (Schery et al. 1980).

The two-filter tube can be used to sample the air intermittently or continuously.

The following precautions must be observed with this method:

- (a) the inlet filter must be well sealed around its edges. An O-ring seal behind the filter is usually needed.
- (b) unused filters in the mining environment can become contaminated. Contamination can be avoided by keeping filters enclosed before and after sampling and by handling them with forceps.
- (c) the calibration of the air-flow meter must take into account the combined resistance of the two filters in series. This can readily be done experimentally with a wet test meter.
- (d) sampling and counting times must be measured accurately.
- (e) when used for thoron gas the transit time of air through the tube must be kept to less than approximately 6 secs. due to the short life of thoron (55 secs.), necessitating larger pumps.

Passive Radon Monitor

In this class of monitor, air containing radon diffuses through porous foam walls into an enclosure of defined volume. Within the enclosure, short-lived daughter products which are produced carrying a positive charge are concentrated onto a small foil by electrostatic collection using a voltage of 1000 volts. A TLD detector located directly behind the foil records the daughter product alpha activity. The monitor is intended to measure the average radon concentrations over periods of a week or longer. These devices use TLD ribbons (thin chips usually $\text{CaF}_2:\text{Dy}$) or track-etch films as the sensing material (Cowper and Simpson 1979). The lowest level of detection is governed by the natural gamma ray background in the area where the monitor is placed since the TLD materials are also sensitive to gamma radiation. An average radon concentration of 1.5×10^{-3} Bq/l (0.04 pCi/l) gives an output equal to that produced by a natural radiation background of 2mR per week. Thus, with a "normal" gamma radiation background of about 2mR/week, exposure to an average radon concentration of 0.1pCi/l should yield a TLD output about 25 times that due to background radiation. The "noise output" of the TLD reader indicated by reading unexposed ribbons is negligible. A second TLD ribbon is used to monitor the gamma background.

The collection efficiency of the positively charged radon daughter product ions depends upon their mobilities which are reduced by the aggregation of water molecules around the ions in a humid atmosphere. Beds of silica - gel are used to make these devices independent of the moisture content of the air.

Passive radon monitors using TLD materials are designed primarily to measure ambient radon levels within buildings, and do not have the sensitivity to measure normal environmental levels which are typically about a factor of ten lower (4×10^{-4} Bq/l). Nevertheless some operators have installed these monitors for environmental surveillance, generally with unsatisfactory results. Laboratory and field trials at ARL have shown a wide and inexplicable variation in sensitivity and suggest that substantial improvements in design are needed before they can be recommended as a reliable instrument.

Radon Passive Track-Etch Detectors

The passive track-etch detector system is attractive as no pump is needed and this lowers the cost significantly. Different types of substrates have been used such as cellulose nitrate film, makrofol and CR-39 polycarbonate. Track-etch detectors were developed for radon detection in the exploration of uranium and have been used in exploration in Australia. Although not effective in finding ore bodies, they have met with some success in detecting radon.

Various authors have been treating the detectors before exposure to lower the background so that occupational levels can be monitored. Hassib (1978) and Massera et al. (1980) use an electrochemical pre-etching technique to prepare an unmarked surface. The electrochemical pre-etching technique may be used to discriminate between a single alpha particle which enters the detector directly from ambient air and those alpha particle clusters which are the result of multiple alpha decay emitted from aerosols deposited on the detector surface. Calibration in radon atmospheres of 4.04 nCi/l where the equilibrium factor F (see section 5) equals 0.5 show a lowest detectable level of 6 WL hrs. For ventilated underground mines, with an equilibrium factor of 0.1 - 0.5 (Domanski et al. 1979) and an exposure time of 3 months, this corresponds to 2.7 - 0.5 pCi/l (0.1 - 0.02 Bq/l).

Track-etch detectors can become electrostatically charged and attract radon daughters to the surface. Recent work, by Chrusciewski et al. (1980) has shown that removal of electric charge from the surface of the film is necessary for reproducible results. Electrostatic negative charges on films produce calibration results that are dependent on the equilibrium factor. A major problem is the calibration of the system against a standard radon/daughter atmosphere taking into account the real aerosol concentration in ambient air (see below). Moreover these detectors are subject to several variables which cannot be controlled namely humidity, temperature, fading and particle concentration.

MEASUREMENT OF AIRBORNE RADON/RADON DAUGHTER CONCENTRATIONS GROWTH OF RADON DAUGHTERS IN AIR

The growth of short-lived radon daughters in air from initially pure radon (i.e. daughter free) can be expressed by the following three differential equations.

$$\frac{dC_A}{dt} = -\lambda_A C_A + C_{RN} \lambda_A \quad (1)$$

$$\frac{dC_B}{dt} = C_A \lambda_B - C_B \lambda_B \quad (2)$$

$$\frac{dC_C}{dt} = C_B \lambda_C - C_C \lambda_C \quad (3)$$

where C_{RN} is the radon gas concentration in air, assumed to be constant and C_A , C_B and C_C are the radon daughter concentrations in air in Bq/l. λ_A , λ_B and λ_C are the decay constants of (^{218}Po) Radium-A, (^{214}Pb) Radium-B and (^{214}Bi) Radium C respectively. ($\lambda_A = 0.227260 \text{ min}^{-1}$, $\lambda_B = 0.025864 \text{ min}^{-1}$, $\lambda_C = 0.035185 \text{ min}^{-1}$.)

t is the build-up time in air in minutes or the "age" of the air.

The solutions to the above equations are:

$$C_A = C_{RN} [1 - e^{-\lambda_A t}] \quad (4)$$

$$C_B = C_{RN} \left[1 - \frac{\lambda_B}{(\lambda_B - \lambda_A)} e^{-\lambda_A t} - \frac{\lambda_A}{(\lambda_A - \lambda_B)} e^{-\lambda_B t} \right] \quad (5)$$

$$C_C = C_{RN} \left[1 - \frac{\lambda_B \lambda_C}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)} e^{-\lambda_A t} - \frac{\lambda_A \lambda_C}{(\lambda_A - \lambda_B)(\lambda_C - \lambda_B)} e^{-\lambda_B t} - \frac{\lambda_A \lambda_B}{(\lambda_A - \lambda_C)(\lambda_B - \lambda_C)} e^{-\lambda_C t} \right] \quad (6)$$

The Working Level is defined as any combination of short-lived decay products of radon (Radium A, Radium B, Radium C and Radium C') in one litre of air which will result in the ultimate emission by them of 1.3×10^5 MeV of alpha particle energy. A measure of the radon activity in air is not an index of hazard of exposure unless the state of equilibrium with its decay products is also known. In some underground mines overseas, where the equilibrium factor is well known, radon concentrations alone are measured. In other countries, an equilibrium factor of 0.4 (Domanski 1979) is quite often assumed to estimate the Working Level. Working Levels determined in this way tend to overestimate the airborne daughter concentrations. Moreover the state of equilibrium frequently differs at different locations in the mine due to ventilation conditions, and is also dependent on the nature of the work being carried out in the mine. Therefore, the best and simplest approach is to measure the daughter concentrations in terms of Working Level directly.

The ingrowth time of radon daughters in terms of Working Level is depicted in Figure 1 but this is not the full story. There can be mixing of "old air", (that is near equilibrium air) with "young air", (far from equilibrium). This mixing can produce a number of daughter ratios not found on the growth curve. This has been illustrated by Rolle 1972 and is shown as Figure 2 where daughter ratios fall within these two curves. Radon daughter ratios in modern uranium mines have been plotted from George et al. (1972, 1977). ARL experience has been that points lying outside these two curves are usually due to measurement errors.

Once formed, a radon daughter atom, which is generally formed as a positive ion may attach itself to an aerosol particle, "plate out" onto any available surface, or survive for some time as an unattached daughter. Because of its short half-life $k_a A$ is more likely to survive its lifetime unattached and hence is often found to have a significant unattached

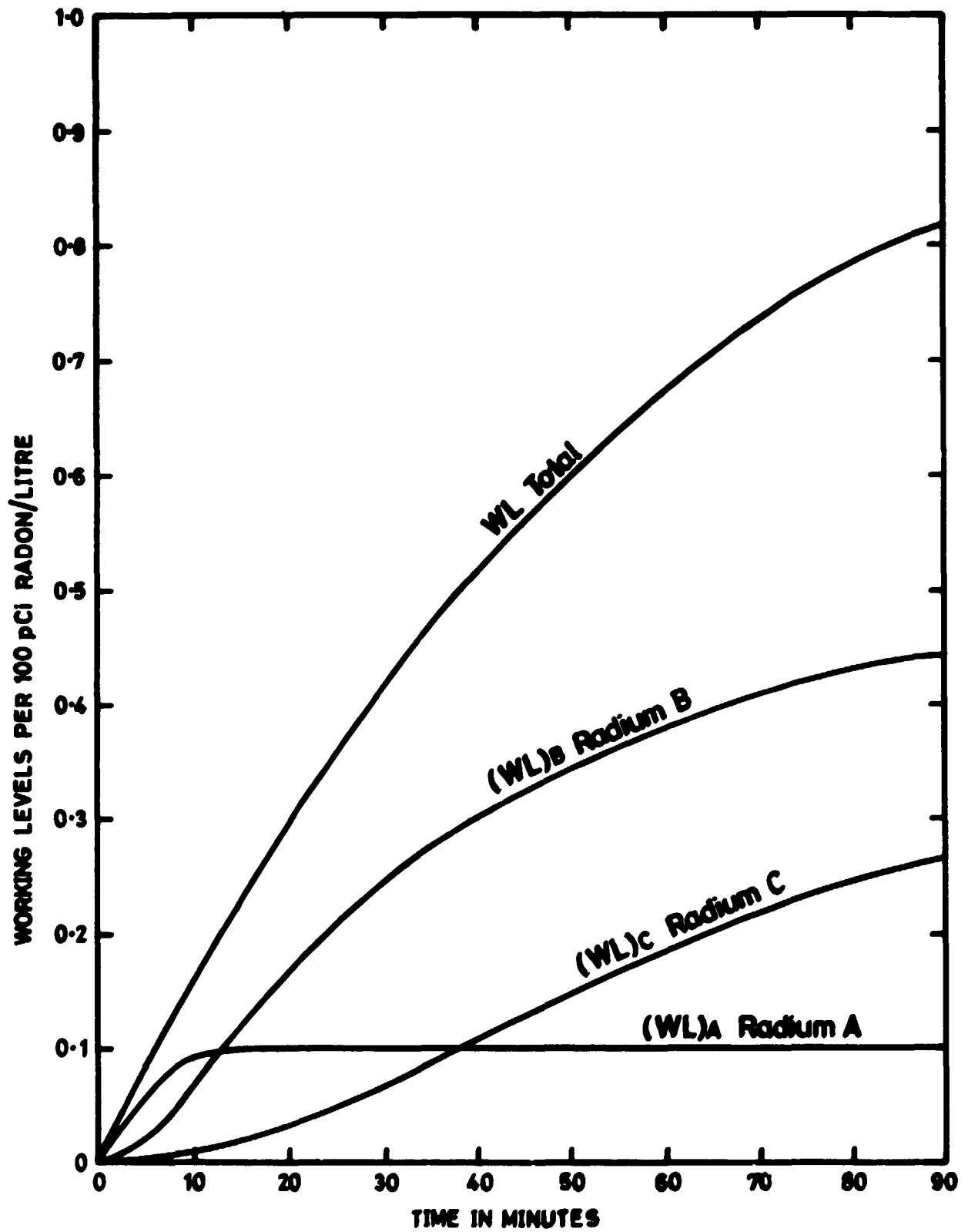


Figure 1. Growth of the total Working Level and the Working Levels of radon daughter as a function of time.

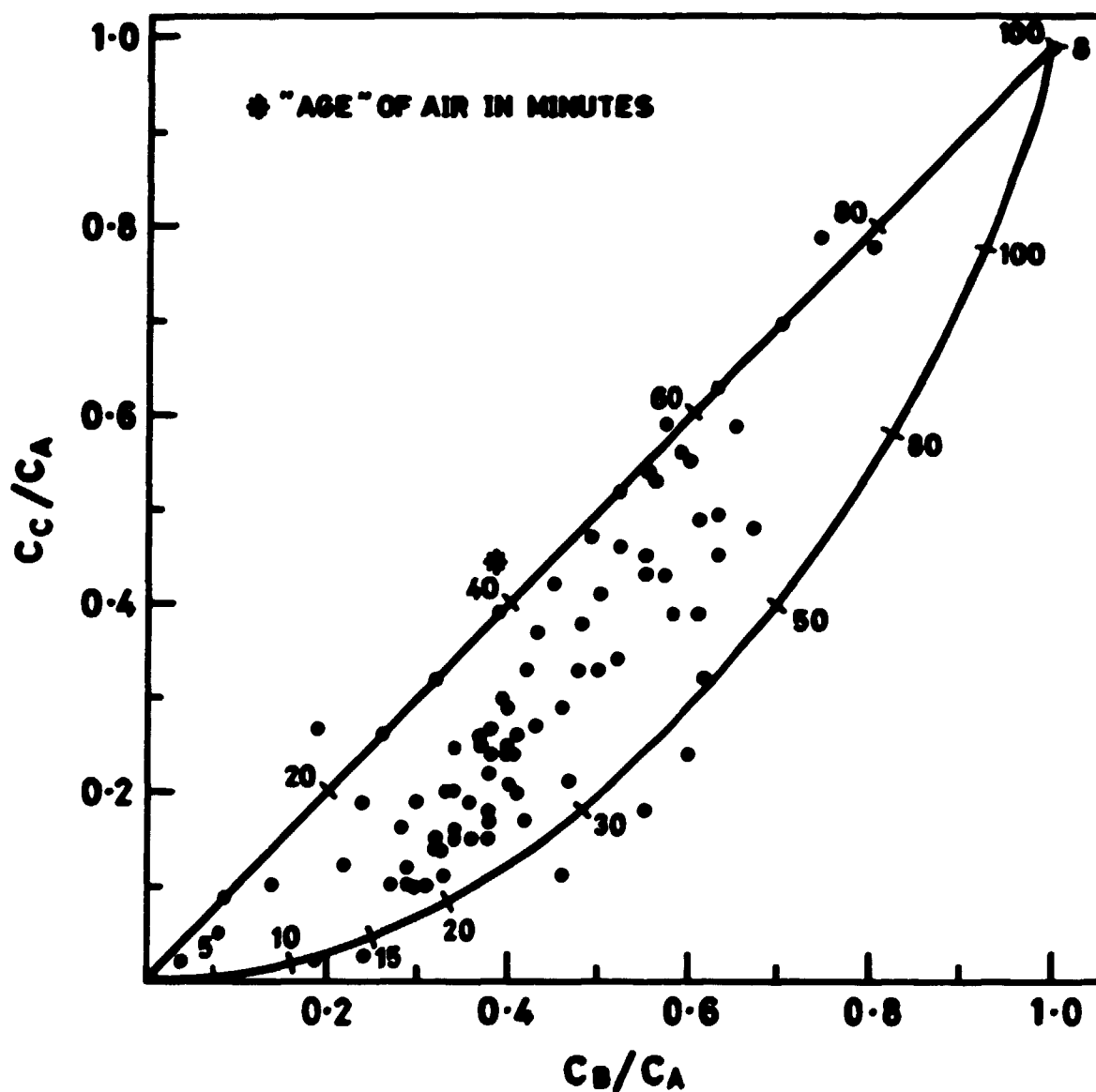


Figure 2. Range of allowed radon daughter concentration ratios when "old" and "new" air is mixed. The curved line represents the activity ratios when initially pure radon decays (i.e. no mixing). The straight line represents the ratios in the limiting case of mixing of old and young air. The experimental points are the results of measurements in mines (George et al. 1972, 1977).

fraction. Not surprisingly, the unattached fraction depends very much on the concentration of particles in the air, and high particle concentrations give rise to low unattached fractions (Duggan and Howell 1969). In underground diesel mines the unattached fraction typically lies in the range 5 - 10%. However in open cut mines, the unattached fraction for RaA can be very much higher, particularly under conditions of good dust control, and we have observed values in the range 20 - 70%.

Plate out of daughters also occurs preferentially to negatively charged surfaces, such as plastic materials (Chrusciewski 1980) and advantage has been taken of this phenomenon to filter out radon daughters from mine air by passing it through beds of small foam plastic beads. Air mixing also appears to enhance plate-out and this method has been used to lower daughter concentrations in some mines (Kaufman and Dinwiddie 1971).

THE MEASUREMENT OF RADON DAUGHTERS

The measurement of radon daughters invariably makes use of the fact that alpha particles are emitted in the decay processes, and these are very easy to detect and discriminate from background. All methods of assessing radon daughter levels involve the filtering of daughters from a known volume of air, and the measurement of the alpha particle activity on the filter paper after sampling. The actual activity measured on the filter paper depends not only on the concentrations of each daughter in the air, but also on the length of time that the air is sampled - this determines how much of the filtered products decay through to RaC. Furthermore, the measured activity depends also on the delay between sampling and counting as the daughters continue to decay. The build up of radon daughters on filter materials (assumed to be 100% efficient) during sampling may be expressed by the set of equations

$$\frac{\partial I_A}{\partial t} = -\lambda_A I_A + QC_A \quad (7)$$

$$\frac{\partial I_B}{\partial t} = -\lambda_B I_B + \lambda_B I_A + QC_B \quad (8)$$

$$\frac{\partial I_C}{\partial t} = -\lambda_C I_C + \lambda_C I_B + QC_C \quad (9)$$

where I_A , I_B , I_C represent the activities of RaA, RaB and RaC respectively on the filter and Q is the sampling flow rate. The above equations assume that the radon concentration and radon daughter concentrations are constant during sampling. The same equations, but without the source terms QC_A , QC_B and QC_C represent the decay of activity on the filter paper at the end of sampling. The solution of these equations, for the two alpha activities $I_A(t)$ and $I_C(t)$, at time t after a sampling interval t_s is straightforward but intricate and is given by

$$I_A(t) = \frac{Q C_A}{\lambda_A} (1 - e^{-\lambda_A t_s}) e^{-\lambda_A t} \quad (10)$$

$$\begin{aligned}
 I_C(t) = Q e^{-\lambda_C t} & \left[\frac{C_C}{\lambda_C} (1 - e^{-\lambda_C t_s}) + \frac{C_B}{\lambda_B} \left(1 - \frac{\lambda_C e^{-\lambda_B t_s}}{(\lambda_C - \lambda_B)} \right. \right. \\
 & - \left. \left. \frac{\lambda_B e^{-\lambda_C t_s}}{(\lambda_B - \lambda_C)} \right) + \frac{C_A}{\lambda_A} \left(1 - \frac{\lambda_B \lambda_C e^{-\lambda_A t_s}}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)} - \frac{\lambda_A \lambda_C e^{-\lambda_B t_s}}{(\lambda_A - \lambda_B)(\lambda_C - \lambda_B)} \right. \right. \\
 & - \left. \left. \frac{\lambda_A \lambda_B e^{-\lambda_C t_s}}{(\lambda_A - \lambda_C)(\lambda_B - \lambda_C)} \right) + \frac{C_B}{\lambda_B} (1 - e^{-\lambda_B t_s}) + \frac{C_A}{\lambda_A} \left(1 - \frac{\lambda_B}{\lambda_B - \lambda_A} e^{-\lambda_A t_s} \right. \right. \\
 & - \left. \left. \frac{\lambda_A}{\lambda_A - \lambda_B} e^{-\lambda_B t_s} \right) + \frac{\lambda_C}{\lambda_B} \left(\frac{\lambda_B}{\lambda_C - \lambda_B} e^{-\lambda_B t} - \frac{\lambda_B}{\lambda_B - \lambda_C} e^{-\lambda_C t} \right) \right. \\
 & + \frac{C_A}{\lambda_A} (1 - e^{-\lambda_A t_s}) + \frac{\lambda_C}{\lambda_A} \left[\frac{\lambda_A \lambda_B e^{-\lambda_A t}}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)} + \frac{\lambda_A \lambda_B e^{-\lambda_B t}}{(\lambda_A - \lambda_B)(\lambda_C - \lambda_B)} \right. \\
 & \left. \left. + \frac{\lambda_A \lambda_B e^{-\lambda_C t}}{(\lambda_A - \lambda_C)(\lambda_B - \lambda_C)} \right) \right] \quad (11)
 \end{aligned}$$

Thus, we can write for the total alpha activity at time t

$$I(t) = I_A(t) + I_C(t) = a_1 C_A + a_2 C_B + a_3 C_C \quad (12)$$

where a_1 , a_2 and a_3 are time dependent coefficients which depend both on the sampling duration and the time delay before the activity is measured.

The quantity we seek to determine, the radon daughter concentration expressed in Working Level units is related to the concentration of each daughter by the relationship

$$WL = 0.0278 C_A + 0.1370 C_B + 0.1008 C_C \quad (13)$$

and a complete determination requires separate knowledge of C_A , C_B and C_C .

The contribution from each daughter obviously depends on the relative proportions of each, usually expressed as the ratio $1 : C_B/C_A : C_C/C_A$, and this in turn depends on the "age" of the air. At one extreme, very "young" air containing only RaA has a daughter ratio 1:0:0, while at equilibrium the ratio is 1:1:1. Figure 3, (after Rolle 1972) illustrates the way in which the filter activity, measured in disintegrations per minute per litre of air sampled is related to Working Level* as a function of observation time for a variety of radon daughter ratios.

* to convert an activity in disintegrations per minute to WL, divide by the sample volume and the factor f , obtained from Figure 3.

One straightforward but limited method for estimating radon daughters is the Kuznetz method (Kuznetz 1956), in which it is assumed that the daughters are in equilibrium when the sample is taken, and a single measurement of the activity is made sometime between 40 and 90 minutes, when the 1:1:1 curve of Figure 3 is changing only slowly with time. The measurement routine is simple, and has the advantage that sampling and counting times are well separated. This is frequently convenient, as a radiation safety officer can take a number of samples relatively quickly around a mine or mill, and count them later under laboratory conditions. However, the method suffers from the severe deficiency that it is substantially in error for "young" air, and the further disadvantage that much of the activity has decayed by the time the measurement is made.

A rather better method requiring only a single measurement of the filter activity is the Rolfe SCAMP method, modified for use with counting equipment rather than instantaneous ratemeters. Rolfe observed that at a particular time after sampling (6.6 minutes for the example given), all of the curves of Figure 3 lie fairly close together, and the conversion factor varies by no more than $\pm 12\%$, independent of the age of the air. The practical application of this method, again modified for counting methods is described in detail in Leach and Lokan (1979).

At the other extreme, the modified Tzivoglou Method, which requires the counting of the filter three times, completely determines C_A , C_B and C_C and hence the daughter concentration in Working Level units. This method, which is also described in Leach and Lokan (1979), is time consuming, taking about 40 minutes per measurement, but very valuable for detailed checking of the age of the air, and the radon daughter ratios.

Spectroscopic Methods

It was pointed out earlier that energy discrimination offered the possibility of distinguishing between alpha particles from RaA and RaC'. Jonassen and Hayes (1973) have developed a method, employing a high resolution alpha detector, and two counting intervals after sampling. This is potentially a very accurate method, but cannot be used readily under field conditions because it needs highly stable laboratory electronics and good quality vacuum systems.

An interesting variation has been proposed by Groer (1973) and developed commercially as a sophisticated Working Level instrument*. Alpha particles from RaA and RaC' are resolved during the counting period, and at the same time the activity from RaB and RaC is measured by detecting the beta particles which pass through the back of the filter. The beta contribution from RaC is inferred from its alpha activity and the RaB activity obtained by difference. The instrument is complex, but can provide a wealth of information on daughter concentrations and daughter ratios as well as a measurement of the Working Level.

A number of other methods have been developed (Hill 1975, Miller 1976, Schiager 1977, James and Strong 1973) which make particular approximations to limit the complexity of counting, or choose particular combinations of alpha and beta detection. Their relative strengths and weaknesses are reviewed elsewhere (Holub and Drouillard 1977, Johnson 1978).

* Harshaw Environmental Working Level Meter.

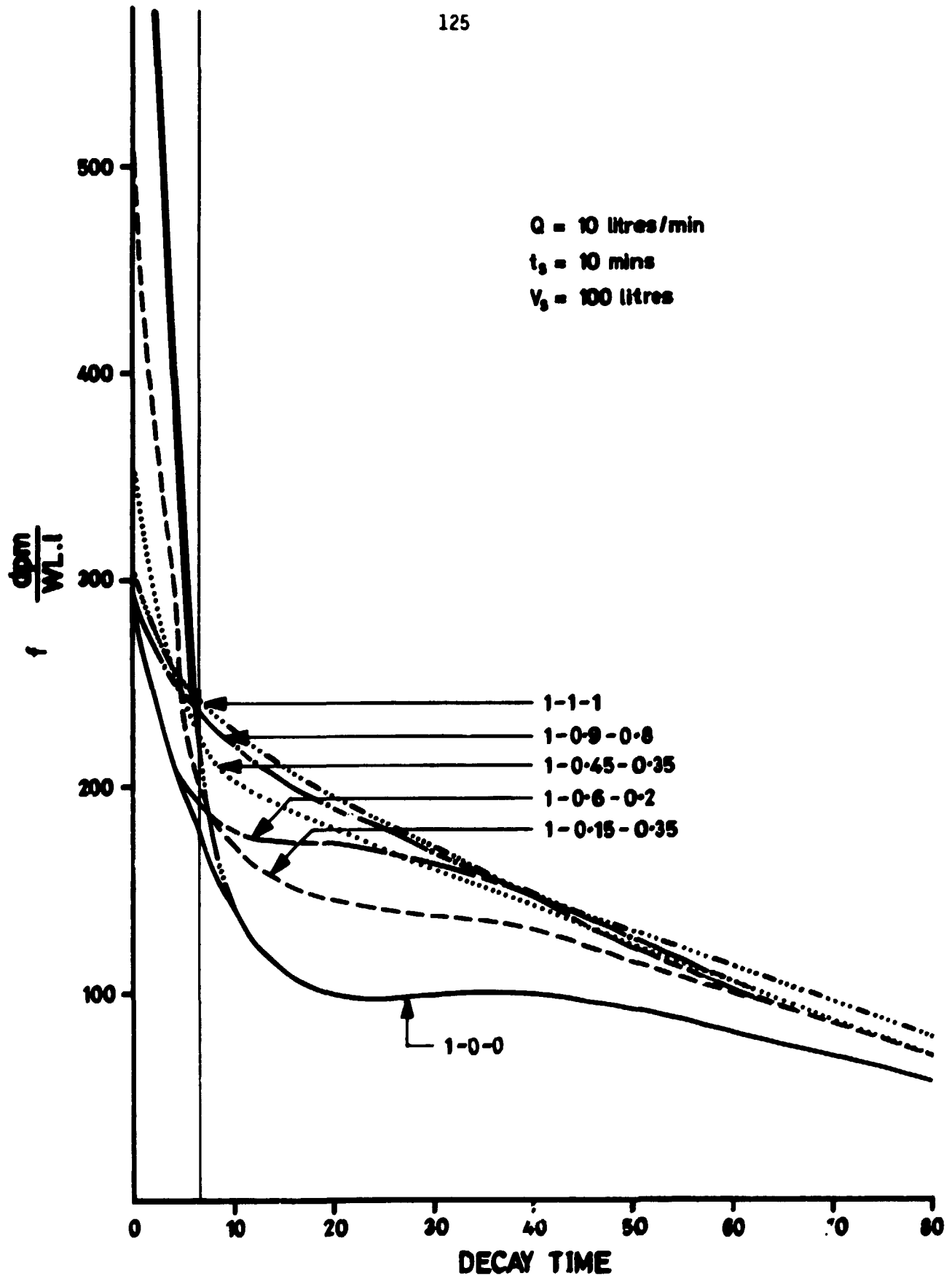


Figure 3. Variation of filter activity with time for various radon daughter concentration ratios (Rolle method - see text).

Personal Radon Daughter Dosimeters

Ideally, employees in uranium mines should have available personal air sampling monitors which measure their individual integrated radon daughter exposures. Several such dosimeters are under development (Phillips et al. 1978, McCurdy et al. 1969), but are not yet in wide use. In general a personal dosimeter takes the form of a small air sampling pump and filter assembly, which is worn by the individual and which filters a known quantity of air during a working shift. Radon daughters are trapped on the filter, and impinge upon a nearby alpha sensitive detector (either a piece of track etch film or a thermoluminescent chip) which is subsequently removed and its alpha particle exposure assessed. Track etch film has the merit that it discriminates clearly between alpha particles and gamma radiation but the disadvantages that it is time consuming to read, and requires a scrupulously consistent etching procedure to ensure a constant sensitivity. TLD chips on the other hand, while easy to read, are sensitive to gamma radiation and the alpha particle exposure can be obtained only by taking the difference between the alpha- and gamma- exposed chip and a second one which senses only gamma radiation. A prototype dosimeter of this type, using $\text{CaSO}_4:\text{Dy}$ as its TLD material has been developed at ARL, and appears to perform satisfactorily under field conditions. Its lowest detectable exposure is about 1 WL hour at an air sampling rate of 0.5 l/min.

Recently Durkin (1979) has described a dosimeter which detects alpha particles from the filter with a surface barrier detector and determines the integrated exposure by conventional pulse height analysis and pulse counting. While this approach promises a higher sensitivity it may prove to be less robust under field conditions than the track etch or TLD detectors.

The WL for radon and thoron must be determined separately. The maximum permissible exposure for radon daughters is 4 WLM per year and thoron daughters is 40 WLM per year. It is prudent to note that radon daughter dosimeters, using alpha energy resolution, can be interfered with by the presence of thoron daughters in air. Thus, the working Level for radon and thoron daughters must be determined separately.

THORON DAUGHTER MONITORING

The monitoring of thoron daughters turns out to be rather simpler in practice than radon daughters as the first daughter with a long enough half life to form significant accumulations is ThB (10.6 hours) (cf Tables 2 and 3). Although not an alpha emitter itself, it is the most abundant source of atoms available for decay though ThC and ThC'. Because of the longer half-life of ThB, thoron daughters are fairly easy to control, and one air change per 10 minutes limits the thoron daughter alpha particle energy to approximately 10% of its potential. By contrast the same ventilation rate only limits the radon alpha energy to 18% of its potential. Briefly, the relatively long half life of ThB leads to another important consequence, namely that since the lung clearance time is only a few hours (Jacobi 1964) a smaller fraction of the total alpha particle energy is deposited in the lung, and the corresponding maximum permitted limits of exposure to thoron daughters are less stringent. While it is likely to be revised downwards in the future (because of an improved understanding of lung dynamics), the present maximum permitted exposure to thoron daughters under the Code of Practice is 40 WLM.

The deposition and decay of thoron daughters on a filter paper may be analysed in much the same way as we have done earlier for radon daughters, and may be expressed by the following differential equations:

$$\frac{\partial I_B}{\partial t} = -\lambda_B I_B + Q C_B \quad (14)$$

$$\frac{\partial I_C}{\partial t} = -\lambda_C I_C + \lambda_C I_B + Q C_C \quad (15)$$

where I_B and I_C represent the activities of ThB and (ThC-ThC')*, C_B and C_C are their concentrations in air, and $\lambda_B = 1.086 \times 10^{-3} \text{ min}^{-1}$ $\lambda_C = 1.144 \times 10^{-2} \text{ min}^{-1}$ are the corresponding decay constants. Again, the same equations, without the source terms $Q C_B$, $Q C_C$ describe the subsequent decay.

The solution of these equations is:

$$I_B(t) = \frac{Q C_B}{\lambda_B} (1 - e^{-\lambda_B t_s}) e^{-\lambda_B t} \quad (16)$$

$$I_C(t) = \frac{Q C_B}{\lambda_B} \left[\frac{1 - e^{-\lambda_C t_s}}{\lambda_C - \lambda_B} - \frac{\lambda_B e^{-\lambda_C t_s}}{\lambda_B - \lambda_C} e^{-\lambda_C t} + \frac{Q C_B (1 - e^{-\lambda_B t_s})}{\lambda_B} \right] \\ + \frac{\lambda_C}{\lambda_C - \lambda_B} e^{-\lambda_B t} - e^{-\lambda_C t} + \frac{Q C_C}{\lambda_C} (1 - e^{-\lambda_C t_s}) e^{-\lambda_C t} \quad (17)$$

Again, alpha counting is the preferred technique with I_C being the measured quantity. Two measurements of I_C are sufficient to determine uniquely the quantities C_A and C_B which may then be used to compute the corresponding thoron daughter level in Working Level Units. If we choose measurement times of 150 and 240 minutes and a sample time of 60 minutes, for example, the concentrations of ThB and ThC may be derived from equations (16) and (17) as

$$C_B = \frac{10^{-4}}{Q \epsilon} (-1.99 I_{150} + 5.56 I_{240})$$

$$C_C = \frac{10^{-4}}{Q \epsilon} (33.03 I_{150} - 32.69 I_{240})$$

where I_{150} and I_{240} are the net count rates (cps) at 150 minutes and 240 minutes after sampling. C_B and C_C are expressed in units of Bq/l. ϵ is the efficiency of the counter.

* As the half life of ThC' is short (0.3 μ sec) it may be considered as part of the decay of ThC.

Again, the corresponding thoron daughter concentrations in Working Level units may be expressed as the sum of alpha particle energy contributions from each isotope, and given by:

$$(WL)_{Th} = 3.289 C_B + 0.3114 C_C$$

where C_B and C_C are the respective concentrations of ThB and ThC in Bq/litre.

A somewhat simpler field technique, in which a single observation is made after 5 hours (i.e. when ThC is in equilibrium with ThB) has been used by Rock (1975), and yields for the thoron daughter working level

$$(WL)_{Th} = \frac{R}{\epsilon VF}$$

where R is the count rate after 5 hours (min^{-1})
 V is the volume sampled (litres)
 ϵ is the efficiency of the detector expressed as a fraction

and F is a time dependent factor, which is illustrated in Figure 4. The factor F is derived for an equilibrium mixture of ThB and ThC (a daughter ratio of 1:1), but, for extreme disequilibrium (1:0) the difference in the calculated Working Level is only 40% because of the dominant contribution from ThB.

However, the Rock method is valid for short sampling intervals only (up to 1 hour), as no allowance is made for thoron daughter decay over longer intervals.

Routine field methods for determining radon and thoron daughter mixtures have not been developed, as the occurrence of comparable quantities of each is very unlikely. However, in some circumstances the presence of modest concentrations of thoron daughters must be allowed for (after separate measurement) when radon daughter levels are being assessed.

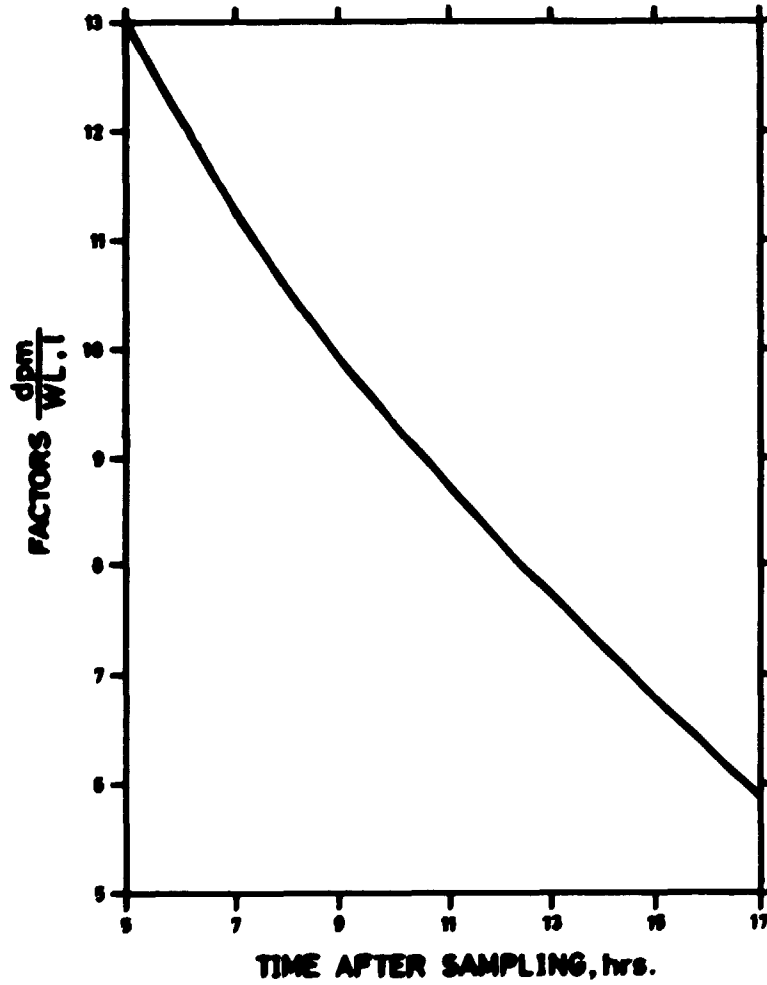


Figure 4. Factors for converting disintegrations per minute per litre of air sampled, 5 to 17 hours after sampling, to Thoron-daughter working levels.

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