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Recommended Procedures for Measuring Radon Fluxes from Disposal Sites of Residual Radioactive Materials

Prepared by J. A. Young, V. W. Thomas, P. O. Jackson

Pacific Northwest Laboratory
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Prepared for
U.S. Nuclear Regulatory
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ABSTRACT

This report recommends instrumentation and methods suitable for measuring radon fluxes emanating from covered disposal sites of residual radioactive materials such as uranium mill tailings. Problems of spatial and temporal variations in radon flux are discussed and the advantages and disadvantages of several instruments are examined. A year-long measurement program and a two month measurement methodology are then presented based on the inherent difficulties of measuring average radon flux over a cover using the recommended instrumentation.

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RECOMMENDED PROCEDURES FOR MEASURING RADON FLUXES FROM DISPOSAL SITES OF RESIDUAL RADIOACTIVE MATERIALS

J. A. Young, V. W. Thomas and P. O. Jackson

I. Introduction

The U. S. Environmental Protection Agency (EPA) interim environmental standards for the disposal of residual radioactive materials from inactive uranium processing sites were published in the Federal Register in January 1981 (40 CFR 192). Although the Department of Energy (DOE) has the primary responsibility for the implementation of these EPA standards, PL-95-604 requires that the Nuclear Regulatory Commission (NRC) concur in remedial actions. One of the requirements of the interim standards is that disposal of residual radioactive materials from inactive uranium processing sites shall be conducted in a way that provides a reasonable expectation that the average annual release of radon-222 from the disposal sites to the atmosphere by residual radioactive materials following disposal will not exceed 2 pCi/m²-sec for at least 1000 years following disposal. However, the EPA has recently proposed that the standard be changed to require that the flux shall not exceed 20 pCi/m²-sec for at least 200 years, and to the extent practicable, 1000 years. It is expected that the radon flux standard will be interpreted to require that the annual average flux from a tailings pile, rather than the flux at any location on the pile, shall not exceed the standard.

It is anticipated that the reduction of radon emissions from disposal sites will be accomplished by covering the tailings with a layer, or layers, of earthen material. However, it is possible that a layer of material such as asphalt will also be laid down to act as a radon barrier. The cover will decrease the emission of radon into the atmosphere because of the radioactive decay of the radon during its diffusion through the cover. The radon emission will also be decreased because the cover will reduce the concentration gradient and therefore the rate of diffusion of radon from the tailings.

The radon flux from a covered tailings pile will come from both the tailings and the covering material. The radon emission standard will be considered to be satisfied for a disposal site if the radon flux is less than or equal to the standard plus the exhalation rate of the cover material. Fluxes from natural soils are typically 0.5 to 1 pCi/m²-sec, but fluxes up to several times these values are not unusual. Therefore, the radon fluxes from possible cover materials at each disposal site should be determined as part of the disposal plan.

The final EPA standard concerning radon fluxes may be in the form of either a performance standard or a design objective. A performance standard would require that radon flux measurements be performed to verify compliance. A design objective would require only that the cover be designed to lower the calculated radon flux below a given value. Flux measurements would not be required to verify compliance with a design objective. However, radon flux measurements would still be useful for the experimental purpose of verifying that the design cover is functioning as planned.

This report has been prepared for NRC to recommend procedures for measuring radon fluxes from disposal sites of residual radioactive material after they have been covered to reduce the radon flux. It will recommend sampling programs, instrumentation, analytical procedures, data reporting formats, and statistical analysis of the data that should be used in the determination of radon fluxes.

II. Spatial and Temporal Variations in the Radon Flux

1. Spatial Variations

The determination of average radon fluxes from disposal sites is complicated by the fact that there may be large spatial and temporal variations in the flux from a given disposal site. The flux from a tailings pile varies with location on the pile because of variations in thickness of the pile and variations in the particle size, ^{226}Ra concentration, moisture content, and emanating power of the material added to the pile (the emanating power is the fraction of the radon atoms produced by ^{226}Ra that escapes the crystal lattice and is free to diffuse). Measured radon fluxes have varied by more than an order of magnitude with location on tailings piles (Silker and Heasler, 1979; Ford, Bacon and Davis Utah Inc., 1981).

According to Leggett, et al. (1978), the number of locations at which a parameter must be measured to determine its average value with a precision of 25% at the 95% confidence level is given by

$$\text{Number} = 45(\text{coefficient of variation})^2 \qquad (1)$$

The coefficient of variation of the radon flux measurements made by Silker and Heasler (1979) at several locations on the Grants, New Mexico tailings pile was 0.74. Freeman (1981) found that the coefficient of variation with location was 0.84 for the Grand Junction tailings pile. The coefficients of variation of the radon flux measurements made by Ford, Bacon and Davis Utah Inc. (1981) also averaged 0.84 for several uncovered tailings piles. According to Equation (1), the number of locations at which the flux would have to be measured to determine the average within 25% at the 95% confidence level would be 25 if the coefficient of variation were 0.74, and 32 if it were 0.84.

The variation of the radon flux across a covered tailings pile could be somewhat less than that across an uncovered pile because horizontal diffusion of radon in the cover material would be expected to lower horizontal concentration gradients. However, if the cover material were not uniform, or if cracks developed in it, the spatial variation of the flux from a covered tailings pile could be greater. The coefficient of variation of the radon flux measurements made by Ford, Bacon and Davis Inc. (1981) averaged 0.66 for several tailings piles covered by about six inches of soil. According to Equation (1), measurements at only 20 locations would be required for this coefficient of variation. However, Leggett, et al. (1978) also recommends that measurements be made at a minimum of 30 locations. It therefore appears that in most cases flux measurements should be made at 30 locations, although in some cases measurements of more locations would be required because of higher variations in the radon flux.

2. Temporal Variations

A. Introduction

The radon flux from a given location at a disposal site will also show considerable variation with time as a result of changes in meteorological conditions, moisture content of the tailings, and perhaps settling of the cover material. According to Baver (1956), the meteorological factors influencing the radon flux are, in order of decreasing importance, rainfall, variations in barometric pressure, variation of soil and atmospheric temperature, and wind speed. He estimated that together these factors are responsible for less than 10% of normal soil aeration.

B. Moisture Content

The radon flux will depend greatly upon the moisture content of the tailings and cover material. The fraction of radon atoms produced by ^{226}Ra decay that escape the crystal lattice increases with moisture content. When a ^{226}Ra atom decays by alpha particle emission the radon atom that is formed recoils in a direction opposite from that taken by the alpha particle. If the recoiling atom comes to rest inside a grain of the material, it is very likely to remain entrapped, but if it comes to rest in a pore it will be free to diffuse into the atmosphere. However, the pores of compacted natural materials are likely to be smaller than the recoil range of radon atoms in a gas, so a recoiling atom that enters a gas-filled pore is very likely to cross the pore and become entrapped in a neighboring grain (Tanner, 1980). The recoil range in water is about one hundred times less than that in air, so the probability that a recoiling atom will stop in a pore is greatly enhanced if the pore is water-filled.

The rate of diffusion in water is much less than that in air so the rate of diffusion into the atmosphere of the radon atoms that have escaped the crystal lattice will be lowered by increasing the moisture content of either the tailings or the cover material. Therefore, increased moisture content could either raise or lower the radon flux. According to Momeni, et al. (1979), the radon flux from domestic uranium ores varies only slightly with moisture content between 10% and 80% saturation. On the other hand, Rogers, et al. (1979) found that the flux from tailings decreased by a factor of 100 when the moisture content increased from dry to saturation. The apparent contradiction of these results is explained by the observations of Strong and Levins (1982), who measured the flux of radon from a column of mill tailings as a function of moisture content. They found that the flux increased by a factor of 3.5 when the moisture content increased from 0.2 to 5.7% by weight. It then increased only slowly with increasing moisture content until saturation was reached, at which time it decreased sharply. They estimated that the radon flux from an infinitely thick tailings pile that was saturated with water would be only 1% of that from a pile containing 5.7% water.

According to Momeni, et al. (1979), a moisture content of 0.2% is typical for dry tailings in a southwestern climate. Increasing the moisture content of these tailings would be expected to increase the flux. However, if the tailings were covered by a layer of earthen material, increasing the moisture content of the cover material would decrease the rate of diffusion of radon through the cover and lower the flux. A heavy rainfall might not immediately increase the moisture content of the tailings, but it would increase the moisture content of the cover material, thereby greatly decreasing the radon flux. Therefore, flux measurements made following a heavy rain should not be used to determine average fluxes.

C. Ice

Several investigators have also observed that a cover of ice will sharply reduce the radon flux from crustal surfaces. Pearson and Jones (1965, 1966) observed that the flux increased by a factor of two or more when a winter thaw resulted in the disappearance of an ice cover. Countess (1977) observed that a 1000-fold reduction in the flux from a tailings pile persisted for several weeks following the formation of an ice cover on the pile. Therefore, the average flux from a tailings pile should not be determined by flux measurements made when the pile is covered with ice.

D. Pressure

Decreasing atmospheric pressure draws interstitial gas toward the surface, thereby increasing the radon flux; and increasing pressure pumps it away from the surface, thereby decreasing the flux. According to Clements and Wilkening (1974) atmospheric pressure changes of 1% or less cause 50 to 100% changes in the exhalation rate of soil, with the actual change depending upon the rate of change of the pressure and the duration of the change. A frequently quoted figure is that a 1% atmospheric pressure change will cause approximately a 60% change in the radon flux (Collé, et al., 1981). Bogoslovskaya, et al. (1932) found that the flux from uranium ore would vary by an order of magnitude with atmospheric pressure, even if the ore were buried five meters below the surface.

E. Wind Speed

It is not known for certain how significant a role wind speed plays in determining radon fluxes. The uncertainty is partly due to the fact that radon flux measuring devices interrupt the flow of air across the material whose flux is being measured. Therefore, it is not possible to be certain that the flux from the material is the same as it would be if the flux measuring device were not there. Pearson and Jones (1966) found no obvious correlation between wind speed and the radon flux from grass-covered soil in Illinois at the very low wind speeds that normally occurred near the soil surface. However, at abnormally high wind velocities the flux increased linearly with velocity. Kraner, et al. (1964) measured higher fluxes on unstable days with higher wind speeds. They postulated that the increase occurred because the microoscillations in barometric pressure that are associated with wind gave rise to turbulent pumping that resulted in the exchange of a layer of soil gas with radon free air from above the surface. Israel and Horbert (1970) measured about a four-fold increase in the radon flux from soil when the wind speed increased from 1 to 13 $msec^{-1}$. However, their measurements were performed on moist soil, and they concluded that the increase in the flux was due to a decrease in the soil moisture content at higher wind speeds. Because of the possibility that radon fluxes increase with increasing wind speed, fluxes should not be measured during high winds.

F. Season

The radon flux can be expected to show seasonal variations at locations that show seasonal variations in factors such as soil moisture or ice cover. Megumi and Mamuro (1973), however, found little seasonal variation at Osaka. Bakulin (1969) found that the seasonal variations were not more than 10%, with maximums occurring in summer. Because of the possibility of systematic seasonal variations, the radon flux should be measured at uniform intervals throughout the course of a year in order to obtain a reliable value for the average flux.

G. Diurnal Variations

The radon flux may be expected to show diurnal variations because of (1) the diurnal pressure wave, which produces a minimum in the pressure in the afternoon; (2) turbulent mixing in the atmosphere which leads to an increase in the flux during the day; and (3) changes in convective flow due to temperature differences in the soil between day and night. Pearson and Jones (1966) found that the flux from soil in Illinois was highest near sunrise and in the mid-afternoon when the atmosphere was most turbulent near the soil surface. The maximum (hourly) radon fluxes during the day were around seven times the fluxes measured during the stable nighttime. Duwe (1976) concluded from a study of the measurements by seven investigators that the most likely pattern of radon flux is a broad nighttime minimum, an increase during the morning to an average value of about 2.5 times the minimum, a decrease during the early afternoon, and a second increase during the late afternoon to an average value of about 1.5 times the minimum. Duwe also concluded from model calculations that soils with low permeability would show lower diurnal flux variations. Because of the likelihood of diurnal variations in the radon flux, radon flux measurements should, if possible, be made over time periods that are multiples of 24 hours.

H. Trends with Time

Radon fluxes from stabilized tailings piles could show systematic trends with time because of factors such as (1) changes in the moisture content of the tailings and cover material, (2) development of fissures in the cover, (3) erosion, (4) the action of burrowing animals, and (5) the growth of vegetation. Changes in the moisture content would be particularly likely to cause significant trends. If the cover material were sprinkled with water during its addition, the radon flux might be expected to increase rapidly with time at first, and then change more slowly after that as equilibrium moisture content was approached. This author is not aware of any available data that could be used to determine the time period required for moisture content and radon fluxes to approach equilibrium levels. The time required would depend upon the climate and the nature of the cover material. Therefore, radon flux measurements at a few locations on at least two tailings piles should be made at least once a month for about a year to determine the time that should be allowed for the fluxes to approach equilibrium values before extensive measurements to determine the average flux are initiated. After the fluxes have approached equilibrium values on these two piles, the flux measurements should be continued on a once a year sampling schedule for as long as possible to determine the nature of any long-term trends.

III. Radon Flux Measurement Techniques

1. Introduction

Several investigators have used various types of accumulators or charcoal canisters to measure radon fluxes. However, at the present time there exists no facility that can be used to accurately calibrate these flux measuring devices under varying meteorological conditions. The flux is generally calculated by dividing the total quantity of radon collected in the device by the area covered by the device and by the sampling time. Therefore, it is not really possible at the present time to compare the accuracies with which the various devices measure the radon flux.

2. The Charcoal Canister Method

Several investigators have employed various types of passive charcoal canisters to measure the radon flux. The canister containing charcoal is placed directly in contact with the surface. The charcoal adsorbs the emanating radon, and after a period of time ranging from a few hours to a few days the charcoal is removed and the average flux determined from the quantity of radon adsorbed on the charcoal. The radon is usually measured by sealing the charcoal in an air tight container, allowing the charcoal to sit for a few hours to allow the short-lived radon daughters to come to equilibrium with the radon, and counting the gamma-rays emitted by the short-lived radon daughter, ^{214}Bi, using either a NaI(Tl) or a germanium diode gamma-ray spectrometer. However, the radon can also be desorbed from the the charcoal and counted in a ZnS scintillation detector cell. The charcoal canister method has the advantage that many measurements can be made inexpensively because of the low cost of the canisters and the ease with which they can be deployed and recovered.

Countess (1977) has used a modified U.S. Army M11 gas mask charcoal canister to measure radon fluxes. This canister covers an area of 87 cm^2 and contains 148 g of activated charcoal. Countess (1977) reports that a lower limit of detection of 0.03 pCi/m^2-sec can be obtained for a four-day exposure using this canister. This detection limit should be more than adequate for determining whether the flux is greater than 2 pCi/m^2-sec. Mine Safety Appliance Co. manufactures an activated charcoal cartridge type GMA No. 459315 that is suitable for measuring radon fluxes. It will cover an area of 41 cm^2 and contains 36 g of charcoal (Countess, 1977). It is also quite easy to construct charcoal canisters using PVC pipe or similar material.

MacBeth, et al. (1978) reported that the precision and accuracy of the charcoal canister method is ±15%. This figure may be optimistic, however, because a two-laboratory comparison study performed to determine whether the actual analysis of the charcoal canister is a major contributor to variations in measured fluxes found that the average difference in the measurements between the two laboratories was 16% (Horton, 1979). However, with careful counter calibration it should be possible to measure the radon with a considerably better precision than this.

Charcoal canisters have the drawback that they can only be used to measure the flux over a very limited area for a limited period of time. Therefore, they should be used to measure the flux at several locations and at several times at each location to determine the average radon flux.

Magumi and Mamuro (1972) increased the measurement area to 2,450 cm^2 by spreading the charcoal over a netting laid on the ground. The charcoal was isolated from the atmosphere by covering it with PVC film. Kisieleski, et al. (1980) increased the area measured by attaching an army gas mask canister to the center of a collector lid covering an area of 2,300 cm^2. However, the diffusion of the radon under the collector lid to the charcoal canister may be too slow to prevent the radon concentration under the lid from rising to the level at which it lowers the net radon flux from the emanating surface, so this method could give results that are too low. Therefore, it should not be used to measure radon fluxes until it can be proved to provide accurate measurements. Any time the charcoal canister method is used, care should be taken to minimize the distance between the charcoal and the emanating surface to prevent the radon concentration from building up above the emanating surface.

3. The Flow Method

Several investigators have measured the average radon flux over a relatively large area by circulating the air under a collector through a charcoal bed. Pacific Northwest Laboratory (PNL) developed a recirculating, pressure balanced, flow-through radon flux measuring system that uses a 76 X 122 X 5 cm (9300 cm^2 area) aluminum tent to cover the area to be measured (Thomas, et al., 1982; Freeman, 1981). A diaphragm vacuum pump draws air through a drierite column to remove water vapor, through a filter to remove particulates, and then through an activated carbon trap to remove radon. The carbon trap consists of a 4.8 cm diameter convoluted tube that is filled with 400g of Pittsburgh Carbon Company 8-12 mesh activated carbon. This trap has been shown to absorb 99.9% of the radon in air that is circulating through the trap at a rate of 2 liters per minute at a temperature of 44°C (Hartley, et al., 1981). This sytem is sealed to tailings by pushing the lip of the tent into the tailings. It is sealed to asphalt by means of caulking compound. After about four hours of sampling, the charcoal is transferred to a petri dish and counted after a few hours delay for ^{214}Bi using either a NaI(Tl) or an intrinsic germanium gamma-ray spectrometer to obtain the radon concentration.

The coefficient of variation of the radon flux across the area covered by the PNL flux measuring system is expected to be much less than the coefficient of variation between the fluxes at widely separated locations on the tailings pile. Freeman (1981) found that the coefficient of variation of the fluxes measured at different locations on the Grand Junction tailings pile using the PNL system was 0.84. This is much larger than the coefficient of variation of 0.29 that Silker and Heasler (1981) measured between four locations within an area of 200 cm^2 using a 41 cm^2 area charcoal canister. Countess (1977) found an even smaller coefficient of variation between multiple measurements of radon flux over a one to two square meter area on several test surfaces. He found that the coefficient of variation ranged from 0.06 for an outdoor location in the phosphate region of Florida to 0.15 for measurements on soil in New Jersey. The variation in the flux across a covered tailings pile will be dependent upon the degree of heterogeneity of the tailings and cover material. However, if it is assumed that the coefficient of variation of the flux (as measured by a charcoal canister) across the PNL system will be 0.29, and the coefficient of variation in the flux (as measured by the PNL system) across the entire tailings pile will be 0.84, then it can be calculated that using a charcoal canister rather than the PNL system will only increase the coefficient of variation of the measured fluxes from 0.84 to $(0.84^2 + 0.29^2)^{\frac{1}{2}} = 0.89$. According to Equation (1), this would only increase from 32 to 36 the number of locations at which it was necessary to measure the flux in order to determine the average flux with a 25% accuracy at the 95% confidence level. It therefore appears that the average flux over a large area could be determined just as accurately with a charcoal canister as with the PNL system, although a few more measurements might be required. It should be remembered also that a charcoal canister can be used to measure the radon flux over a longer time period than can the flow system, so a single measurement using a charcoal canister would probably provide a better estimate of the temporal average than would a single measurement using a flow system. In summary, comparisons between a charcoal canister system and a flow system indicate that charcoal canisters are more effective in terms of cost and effort for measuring the average radon flux across a large area such as a reclaimed disposal site. However, the accuracies of the two techniques must still be compared using a calibration facility before a choice can be made between them.

4. The Accumulation Method

The accumulation method involves the measurement of the radon that accumulates in an open-faced container that is inverted and sealed to the emanating surface. The accumulator is generally sealed to a soil surface using wet bentonite or by imbedding the rim of the accumulator several centimeters into the soil. The accumulator is sealed to rigid surfaces such as building materials using epoxy resins or other caulking agents. Accumulators of many sizes and shapes have been used, with large barrel accumulators being popular.

The radon flux is determined by measuring the initial rate of change in the radon concentrations in samples of air that are withdrawn periodically from the accumulator through a sampling port. The air in the accumulator is generally mixed with a small fan to insure that representative samples are obtained. The flux is calculated using the equation

$$E = \frac{V}{A}\left(\frac{\Delta n}{\Delta t} + \lambda \bar{n}\right) \qquad (2)$$

where E = radon flux (atoms/cm^2-sec)
V = volume of accumulator (cm^3)
A = surface area of accumulator (cm^2)
n = radon concentration (atoms/cm^3)
t = time (sec)
λ = radon decay constant (sec^{-1})

The rate of change in the radon concentration in the accumulator can be used to calculate the radon flux only until such time as the concentration reaches a level that is a significant fraction of the concentration in the emanating material. At that time back diffusion into the emanating material will decrease the concentration gradient in the emanating material and thereby lower the net flux into the accumulator. Wilkening, et al. (1972) recommends that the concentration in the accumulator be kept below 10% of the soil gas concentration at a depth of 13 cm. For most soils this concentration is reached in a matter of hours.

Errors may arise in the measurement of radon fluxes using accumulators because of errors in the measurement of the quantity of radon in the accumulator, and because the accumulator (1) changes the flux by disturbing the soil, (2) changes the soil temperature, which may change the thermal stability or the amount of radon adsorbed onto soil grains, (3) reduces the flux because of increased radon concentrations inside the accumulator and (4) changes the temperature, wind velocity, and turbulence above the soil surface (Duwe, 1976). However, the same difficulties are faced by the charcoal canister and flow methods.

Wilkening (1977) reported that typical error limits for the accumulation method are 6 to 10%. Bernhardt, et al. (1975) performed the most extensive evaluation and verification of the accumulation method. They found that although the counting errors were generally less than 5% for each radon sample, the precision for replicate flux measurements was typically 20% for fluxes of 100 pCi/m^2-sec and 50 to 100% for fluxes of less than 10 pCi/m^2-sec.

The accumulator has the advantage that it can be used to measure the radon flux over a larger area than is generally measured using a charcoal canister. However, sampling time is limited because of the build-up of radon in the accumulator. The accumulator is a much more complicated and expensive device than a charcoal canister, and the measurement of radon is more complicated using the accumulator. It also appears that the precision of accumulator measurements at low radon fluxes is not very good. These factors would seem to indicate that the accumulator method would be a less satisfactory method for conducting radon flux surveys than is the charcoal canister method. However, the accumulator method could still be the method of choice if it could be shown to provide more accurate flux measurements than other techniques.

5. Track Etch® and Thermoluminescent Dosimeter (TLD) Detectors

Radon fluxes show large temporal variations, so average annual fluxes should be determined from several measurements during the course of a year if a measurement technique is used that is not capable of making a measurement over a period of longer than a few days. Therefore, it might seem more practical to measure the radon flux using a Track Etch® or TLD detector which was buried beneath the soil surface, or attached to the surface of a material such as asphalt or concrete, and left in place for a year or more. Extensive measurements of soil gas concentrations have been made using these devices by many investigators, especially during the exploration for uranium deposits. However, the Track Etch® and TLD detectors measure the radon concentration rather than the flux. Therefore, the radon concentrations would have to be measured at several depths and the effective diffusion coefficient determined before fluxes could be calculated from these concentration measurements. Alternatively, it might be possible to derive approximate empirical factors relating single-depth radon concentrations to radon fluxes from simultaneous measurements of concentration and flux for various materials. Wilkening, et al. (1972) found that there was a good correlation between radon flux and soil gas concentration near Socorro, New Mexico. However, the derived factors might be expected to be different for different materials, and might be expected to change with meteorological conditions and soil moisture. Therefore, it appears that the measurement of radon fluxes using Track Etch® or TLD detectors would not be practical until extensive simultaneous measurements of concentrations and fluxes had been made to derive empirical factors relating concentrations to fluxes for various materials and conditions.

IV. Procedures for Conducting Radon Flux Surveys

1. Summary of Recommended Procedures

A gamma-ray survey should be performed using a detector system such as a micro-R-meter to measure the gamma-ray exposure rates at an elevation of 80 to 140 cm at the grid points of 350 by 350 cm grid. If an increase in the exposure rate is detected at any location, a search around that location should be made at the surface for elevated contact readings. Radon fluxes should be measured at locations showing exposure rates greater than three standard deviations above the average for the tailings pile. Flux measurements should also be made at enough locations on a rectangular grid to bring the total number of measurements up to the number required by Equation (1) or to 30, whichever is greater.

Each flux measurement should be made over as long a period of time as is practical, preferably two or three days. The measurements should not be made after a heavy rain, when there is an ice cover, or during high winds. If the cover material has been sprinkled with water during application, then flux measurements should not be begun until the covered tailings pile has dried out enough so that the radon fluxes have stopped increasing rapidly with time. Repeated measurements at a few locations on at least two of the first piles measured should be used to estimate how long a time should be waited. Ideally, the flux measurements should be made every other month over the course of at least one year. However, if the flux measurements are being made to determine whether the flux exceeds a performance standard, it may be necessary to complete the measurements within a shorter period of time, so that a decision can be made as to whether further remedial action is required. In that case, flux measurements should be made once a week for two months at each location.

If the measurements are being made to determine whether the average flux exceeds a performance standard, they should be discontinued whenever it becomes possible to be reasonably certain whether or not the average flux will exceed the standard. The measurements should be discontinued if at any time it is calculated that there is either a less than 5% probability that the average net flux will be greater than the existing flux standard, or a greater than 95% probability that the average net flux will exceed the standard (net flux equals total flux minus the flux from the cover material). After the measurements have been completed, the average and the coefficient of variation of the measured fluxes should be used to calculate the probability that the true average flux exceeds the standard.

On the other hand, if the flux measurements are being made to determine whether the cover is performing as designed, fluxes from at least a few tailings piles should be measured every other month for at least one year, because the fluxes could change systematically with time as a result of factors such as changes in soil moisture, erosion, settling of the cover material, growth of vegetation, and the action of burrowing animals. After the first year the measurements should be made once a year until it is certain that there are no significant long-term trends in the radon fluxes.

2. Gamma-Ray Surveys

Considerably elevated radon fluxes could occur at isolated locations on a covered tailings pile because of (1) fissures in the material used to stabilize the tailings pile, (2) elevated exhalation rates from the underlying tailings material, or (3) variations in the thickness of the stabilizing material. Elevated gamma-ray exposure rates could occur at these locations because of the emission of gamma-rays from radon daughters that would deposit on the cover material. It is quite likely that at least some of these "hot spots" would be missed during a radon flux survey consisting of measurements at 30 or so locations. Therefore, it would be desirable to determine the locations of these hot spots, and to make flux measurements at these locations.

For the above reasons, gamma-ray surveys should be conducted before radon flux measurements are made. The measurements should be made using micro-R-meters at an elevation of about 80 to 140 cm at the grid points of about a 3.5 X 3.5 m grid (Young, et al, 1982). This is a considerably denser grid than is likely to be used for the radon flux measurements. If an increase in the gamma-ray exposure rate is detected at any location, a careful search should be made at the surface around that location for elevated contact exposure rates. The average exposure rate and the coefficient of variation of the exposure rates should then be calculated from the measurements at the grid points. Radon flux measurements should be made at locations showing exposure rates greater than three standard deviations above the average.

It may be that the gamma-ray surveys will detect no significant hot spots. If this is found to be the case for the first few tailings piles measured, then the gamma-ray surveys may be discontinued for subsequent piles.

3. Radon Flux Sampling Grid

According to Leggett, et al. (1978), a parameter should be measured at 30 locations, or at a number of locations equal to 45 times the square of the coefficient of variation of the measurements between the sampling locations, whichever is greater. Therefore, in addition to the flux measurements made at locations of elevated gamma-ray exposure rates, measurements should be made at enough grid points on a rectangular grid to bring the total number of measurements up to at least 30. The coefficient of variation of the measurements should then be calculated to determine from Equation (1) whether additional measurements should be made. If additional measurements are required, they should be made at locations where the original measurements have indicated that elevated radon fluxes might be present. It may turn out to be cost-effective to make more than 30 flux measurements initially to insure that it does not turn out to be necessary to go back later to make additional measurements.

4. Time Schedule of Flux Measurements

A. Year-Long Measurement Series

The radon flux at any location will fluctuate with time as a result of meteorological conditions and the moisture content of the emanating material. Since the fluctuations could have a seasonal component, radon fluxes should be measured every other month throughout at least a year to obtain the annual average. If the measurements are being conducted to determine whether the cover is performing as designed, then measurements should be made once a year after the first year or so until it appears certain that the flux is not changing significantly with time.

Each flux measurement should be made over as long a time period as is practical for the measurement techniques being used. If charcoal canisters are used, each measurement should be made over a period of at least one, and preferably two or three days because of the possibility of diurnal and other short-term variations. It is not practical to sample over much longer time periods than this because radon has only a 3.8 day half-life, so the radon originally collected would mostly decay away before measurement if longer sampling periods were used. Also, the saturation of the charcoal by moisture and radon during longer sampling periods might lower the adsorption efficiency of the charcoal. The adsorption efficiency of the charcoal canister system used should be determined as a function of sampling time by making side-by-side measurements on homogeneous tailings material whose moisture content and radon flux is higher than would be expected for the actual covered tailings piles that are to be measured. The measured radon fluxes for given time periods should then be compared with fluxes measured simultaneously over shorter time periods to determine how long a time it takes for the collection efficiency to begin to decrease. The measurement periods for tailings piles should be kept short compared to this time.

Radon fluxes will be measured during only a small fraction of the total time even with an ambitious measurement program, so the measurements should not be made at times when it is expected that the fluxes will depart considerably from average values. Therefore, measurements should not be made following a heavy rain, when there is ice cover, or during high winds. It is also likely that flux measurements should not be made for a period of time following the completion of the stabilization of the tailings pile. It is probable that the cover material will be sprinkled with water following its placement on the tailings pile so that it can be packed down more readily. If this is done, the radon flux should remain below normal until the cover and tailings material dry out enough to approach equilibrium moisture conditions. Therefore, flux measurements should be made at intervals at a given location on the tailings pile to determine when fluxes appear to stop changing systematically with time. At that time extensive measurements to determine the average flux at a given location from the covered pile may be begun. After measurements have been made on a couple of piles it may be possible to estimate the time it takes fluxes to approach representative values, so that flux measurements may be begun following this delay period on subsequent piles. However, the time required for the cover material to approach equilibrium moisture content could vary greatly with the nature of the cover material and climatic conditions.

The number of measurements that would be required to determine the average flux at a given location with a precision of 25% at the 95% confidence level is given by Equation (1). There have been some repeated measurements at given locations on tailings piles over extended periods of time. On the average, the measurements of Silker and Heasler (1979), Marple and Clements (1977), and Clements, et al. (1978) show a coefficient of variation with time of 0.4. According to Equation (1), six measurements would be required if the measurements showed this coefficient of variation. The variation would be expected to be different at different locations, so the total number of measurements that would be required at any location would have to be determined from the coefficient of variation of the first few measurements at that location.

It may be that the radon fluxes from a given tailings pile will either be so low that it will be clear after a few measurements that the net flux will be less than the radon flux standard, or so high that it will be clear that the average will be greater than the standard. Therefore, if the flux measurements are being conducted to determine whether the average flux exceeds a performance standard, the average and coefficient of variation of the flux should be calculated at each sampling location after the second (and each subsequent) measurement, and then be used to calculate the average and the coefficient of variation of the flux for the total pile. If it is calculated that there is either a less than 5% or a greater than 95% probability that the average flux will exceed the standard, the flux measurements should be discontinued.

After the flux measurements have been completed, the average and coefficient of variation of the measured fluxes should be calculated to determine the probability that the true average flux exceeds the standard. If the average flux exceeds the standard, and the decision is made to add additional cover material to locations showing fluxes greater than the standard, then additional flux measurements should be made at these locations following the addition of the cover material. These measurements should be continued until the probability that the average flux from the pile will exceed the standard is calculated to be less than 5% or greater than 95%, or until the total number of measurements required by Equation (1) is completed.

It is possible that the measurements over an extended period of time will indicate that there is a continued change in the flux with time. In that case, if an extrapolation of the data indicates that net flux could change from less than to greater than the standard (or vice versa) in the future, periodic measurements should be continued, if possible, until it is possible to be reasonably certain whether the final average net flux will be greater than the standard.

The average flux calculated in the above manner will probably be somewhat higher than the true average because sampling locations have been selected where elevated fluxes are expected. The coefficient of variation of the measurements might also be expected to be greater because of this selection of sampling locations, so the number of required sampling locations calculated from Equation (1) would be expected to be greater than would be the case if measurements were made only at grid points. However, there are significant experimental errors in the measurements, and the temporal variations of the radon flux will limit the accuracy of the calculated average fluxes. Therefore, the bias in the calculated average flux caused by the selection of sampling locations should be useful in decreasing the probability that the true average flux will be greater than the standard even though the measured average flux is less than the standard.

The possibility does exist, however, that tailings piles will have a large enough number of small areas of high radon flux (hot spots) to cause the average flux, calculated in the above manner, to exceed the true average flux by an unacceptable amount. Model calculations by Mayer and Zimmerman (1981) indicate that a 1.5 cm diameter hole that extends completely through a 100 cm thick cover will increase the average flux over a 150 cm^2 area by a factor of about 30. Therefore, if a large number of hot spots are detected, the areas of these hot spots should be estimated. Each flux measurement, including the measurements at the grid points, should then be weighted according to the area it represents when the average flux is calculated.

B. Two-Month Measurement Series

It is possible that it will be decided that the requirement of a year-long flux measurement series would produce an unacceptable delay in the verification of compliance with a performance standard. In that case, the flux measurements should be made once a week for two months, even though this shortened measurement schedule would probably result in a decrease in the accuracy of the determination of the average flux. The flux measurements should not be begun until the radon flux has approached equilibrium values. Repeated measurements at a few locations on at least two piles should be used to estimate how long to wait. If the measured fluxes change systematically with time, it may be necessary to continue the measurements, perhaps with lower frequency, until it can be predicted with reasonable certainty whether the average flux will be greater than the standard.

V. Calibration of Radon Flux Measuring Devices

At the present time there exists no standard calibration facility that can be used to calibrate radon flux measuring devices. Such a facility is needed to determine whether these devices are providing accurate measurements of radon fluxes. The following paragraphs will describe the characteristics of a facility that could be constructed and used for calibrating radon flux measuring devices. It is based upon a design proposed by Kearney and Kretz (1981).

The flux calibration facility should be constructed in an air-tight chamber having dimensions of at least 2x2x2 meters. A horizontal, perforated metal plate should be attached to the inside of the chamber about 30 cm above the bottom of the chamber. The plate should extend completely across the chamber. A sheet of porous fabric should be laid on the plate and then covered by a layer of sand or other earthen material having low ^{226}Ra content. A standard NBS ^{226}Ra source should be dissolved in an acid solution and placed in a bubbler in the air space underneath the layer of sand. A pump should be used to recirculate the air underneath the sand bed through the bubbler to transfer the radon produced by ^{226}Ra decay into the air beneath the sand bed. The air should be bubbled through water before being bubbled through the ^{226}Ra solution to prevent the ^{226}Ra solution from evaporating away. After leaving the bubbler the humidity of the air may be reduced by passing it through a dessicant. Care must be taken to insure that the radon becomes well mixed in the space beneath the sand bed, but is not forced up through the bed.

The radon should diffuse up through the sand bed at a constant rate to produce a constant flux of radon from the top of the bed. The radon emanating from the bed should be removed immediately to prevent radon concentrations from increasing above the bed and decreasing the radon flux. Therefore, the air above the bed should be circulated through a charcoal trap that is cooled with dry ice. The circulation system should be designed so that the speed of the air across the surface of the bed can be varied and can be measured.

The radon flux will depend upon the strength of the ^{226}Ra source and the fraction of the radon that decays before it diffuses through the bed. Different source strengths can be used to produce varying radon fluxes. The fraction of the radon that decays before passing through the bed can be calculated from the one-dimensional diffusion equation. It can be shown that for a 10 cm thick layer of sand, more than 95% of the radon should pass through the sand before decaying.

The radon flux through the sand bed can also be determined by measuring the rate at which radon is collected in the charcoal trap through which the air above the bed is being circulated. The quantity of radon collected in the charcoal trap can be measured either by gamma-ray spectrometry or by heating the trap to desorb the charcoal into a ZnS scintillation cell, and measuring the alpha particle emission rate with a photomultiplier. The charcoal trap should be replaced periodically, and the radon concentration measured to determine whether the measured radon flux is constant. If it is not, the reason for the variation must be discovered and eliminated.

The radon fluxes through the sand bed determined by the above two methods should agree within a few percent. If they do not, then the reason for the discrepancy should be determined before the chamber is used for calibrating the flux measuring devices.

It is important to insure that the radon flux through the sand bed does not vary with location on the bed, so care must be taken to insure that the sand has a constant thickness across the bed. The radon flux should be measured at several times and locations across the bed using charcoal cannisters to determine whether there is a spatial or temporal variation in the flux. If there is, the variation could, of course, be due to inaccuracies in the measurements, but if there is no significant variation, then it can be concluded that the flux is constant. If the measured flux does vary, the reason for the variation must be discovered before the chamber is used for calibrating flux measuring devices.

It could be that it will not be possible to obtain sufficiently constant fluxes using the chamber described above. In that case, it may be possible to obtain constant fluxes by placing layers of well-blended mixtures of sand and tailings on the floor of an air-tight chamber. The ratio of sand to tailings can be varied to give radon fluxes covering the range over which the flux measuring devices are expected to be used. The flux, and the temporal variation of the flux, can be measured by circulating the air above the tailings through a charcoal trap that is cooled with dry ice and measuring the rate at which radon is collected in the trap. The temporal and spatial variation in the flux from the bed would be determined by measuring the flux at different times and locations with charcoal cannisters. The objection to this method is that the radon flux could not be related to a standard source of radon.

Once a facility has been constructed that will produce a constant, known radon flux, it should be used to determine the rates at which the various radon flux measuring devices collect radon as a function of the radon flux. The rate of collection should be given by

$$C = AF \tag{3}$$

where C = rate of radon collection (atoms/sec)

A = proportionality constant (cm^2)

and F = radon flux ($atoms/cm^2 \cdot sec$).

It is expected that A will approximately equal the area covered by the device. Each device should be used to measure the radon flux over a range of (1) radon flux, (2) air speed across the surface of the bed, (3) temperature and moisture content of the bed and the air, and (4) pressure to determine the magnitude of the variation of A. If the variation is too great, then the device cannot be used to measure radon fluxes accurately. However, even if A does not change with air speed in the calibration facility, it still may change with wind speed in the field because turbulence may cause changes in the radon flux, and the turbulence spectrum of the atmosphere will not be duplicated in the calibration facility.

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This report recommends instrumentation and methods suitable for measuring radon fluxes emanating from covered disposal sites of residual radioactive materials such as uranium mill tailings. Problems of spatial and temporal variations in radon flux are discussed and the advantages and disadvantages of several instruments are examined. A year-long measurement program and a two month measurement methodology are then presented based on the inherent difficulties of measuring average radon flux over a cover using the recommended instrumentation.

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