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Field Measurements of *In Situ* ^{222}Rn Concentrations in Soil Based on the Prompt Decay of the ^{214}Bi Counting Rate

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Indirect field measurements of the *in situ* ^{222}Rn concentrations in sealed samples of soil have been made based on the prompt decay of the ^{214}Bi counting rate in the 2 h interval immediately following sample collection. Subsequent ^{214}Bi measurements yield estimates of the ^{222}Rn lost during sample collection and the concentration of ^{226}Ra in the samples. These data may be used in the measurement of *in situ* ^{222}Rn concentration gradients, the characterization of the state of ^{226}Ra/^{222}Rn equilibrium in soil samples and calculation of ^{222}Rn surface flux.

INTRODUCTION

Recently, a set of indirect *in situ* ^{222}Rn and ^{226}Ra concentration measurements based on the prompt decay and subsequent build-up, respectively, of ^{214}Pb and ^{214}Bi (half-lives of 26.8 and 19.7 min) was completed on canned samples of soil and uranium mill trailings from the Grand Junction tailings pile, Grand Junction, Colorado.[1] These and earlier prompt ^{214}Bi measurements on the Grand Junction tailings pile[2,3] suggest that on the basis of the prompt decay in the ^{214}Bi γ-counting rate it is possible to estimate the pre-collection ^{214}Bi counting rate in such samples at the time of sample collection, T_0. For this field method, it is assumed that, in the 3–4 h prior to sample collection, the ^{214}Pb and ^{214}Bi in the sample prior to collection are essentially in secular equilibrium with the ^{222}Rn in the sample. Given this assumption, it is possible to indirectly estimate the concentration of the *in situ* ^{222}Rn in the sample at time T_0 and for the 3- to 4-h interval immediately preceding the collection and canning of the sample. This interval is determined by the time required for the ^{214}Pb and ^{214}Bi to come essentially into secular equilibrium with the *in situ* ^{222}Rn in the sample.

Subsequent counting-rate measurements on the ^{214}Bi γ-activity of these same canned samples over a period of approx. 30 days yield a direct measurement of the ^{226}Ra concentration in the samples based on the final, equilibrium, ^{226}Ra/^{222}Rn/^{214}Bi, counting rate. Additional data on the minimum ^{214}Bi counting rate, usually obtained in the period from 4 to 10 h after sample collection, also permit an estimate of the amounts of radon lost in the process of sample collection and canning and serves as a measure of the weakly-held, mobile fraction of ^{222}Rn in the sample at time T_0.

The equilibrium ^{226}Ra/^{214}Bi concentration data combined with the *in situ* ^{222}Rn concentration data at time T_0 are used to determine whether the ^{222}Rn concentration in the sample at the time of collection is (1) in secular equilibrium with the ^{226}Ra in the sample, (2) deficient or has lost ^{222}Rn compared to the final ^{226}Ra/^{222}Rn/^{214}Bi equilibrium value or (3) whether the sample contains an excess of unsupported ^{222}Rn. Unsupported ^{222}Rn is the ^{222}Rn which is introduced into the sample and is not produced by the decay of the ^{226}Ra in the sample. The ^{222}Rn found in excess of the final radon concentration produced by the ^{226}Ra in the sample when it is in secular radioactive equilibrium with its daughter products must be unsupported. However, a sample may have a net deficiency in ^{222}Rn at T_0 and still have received unsupported radon, having lost to its immediate surroundings some of the radon produced by the decay of the ^{226}Ra in the sample as well as a portion of the unsupported radon.

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OBJECTIVES AND TEST-SITE DESCRIPTION

The objectives of these preliminary field measurements of the prompt decay and subsequent build-up of the ^{214}Bi counting rate on soil and tailings samples from the Grand Junction tailings pile were to test the capability of the prompt ^{214}Bi technique to:

1. Measure the concentrations of *in situ* ^{222}Rn and ^{226}Ra in different types of test covers and in the underlying tailings.
2. Evaluate the pre-collection state of equilibrium between ^{226}Ra and ^{222}Rn in these samples.
3. Characterize the source and mobility of the ^{222}Rn in such samples.
4. Develop procedures for evaluating the effectiveness of specific tailings cover designs used to restrict the loss and/or migration of ^{222}Rn from the tailings, and for monitoring the long-term performance of such covers.
5. Provide an additional means of directly studying the field and in the laboratory radon transport mechanisms in soil and uranium mill tailings. With prompt ^{214}Bi measurements it should be possible to evaluate the effects of such soil factors as porosity, permeability, moisture content, as well as changes in meteorological conditions etc. on radon transport.
6. Provide a means of calculating and predicting ^{222}Rn surface flux based on the *in situ* ^{222}Rn and ^{226}Ra concentrations obtained on a set of soil samples collected at different depths from the surface.

Three test areas at the Grand Junction tailings pile were selected for this initial study:

(i) The "Sand Box", a specially prepared 4.6 × 4.6 × 1.8 m deep test area into which six 30.5 cm layers of carefully homogenized tailings had been added. This area was covered by coarse, loose sand.

(ii) The 1979 Asphalt Cover Test Area; an area of the tailings pile covered by an approx. 6–7 cm layer of specially prepared asphalt emulsion which in turn was overlain by a protective layer of adobe clay 17.8–30.5 cm deep. This test area is described in detail by Hartley *et al.*[4]

(iii) The 1981 Barrier Field Test Area; this site included comparative tests of the following different cover systems: an area including multi-layer clay, asphalt emulsion and earthen cover systems. The prompt ^{214}Bi measurements were made only on the uncompacted adobe clay cover, a part of the earthen cover system. This test area is described in detail by Hartley *et al.*[5]

A detailed description of the results of the prompt ^{214}Bi field test and an evaluation of the data for the three test areas noted above is given in a report by Stieff.[1] However, only the data obtained on the uncompacted adobe test site, a part of the earthen cover system, have been summarized and selected for presentation in this paper.

The data for the uncompacted adobe are of particular interest because they demonstrate the unique capability of this new field method to measure the changes in the *in situ* concentration of unsupported ^{222}Rn in soil samples as a function of depth. In addition, and perhaps of equal significance, these data combined with estimates of the soil porosity and moisture content were used to calculate the ^{222}Rn surface flux in pCi/m^2 s at the time the sample was collected. If the data on the minimum ^{214}Bi counting rates (a measure of the mobile ^{222}Rn in the sample) and the ^{226}Ra ^{214}Bi equilibrium counting rates are considered, the potential exists to set both a lower and an upper limit on the expected ^{222}Rn surface flux at the collection site.

SAMPLING AND COUNTING PROCEDURES

The uncompacted adobe clay from the earthen cover test area was collected using a 7.6 cm dia, thin-walled, steel Shelby coring tube with extensions. The soil in between the surface and the sampling interval was removed with an 11.4 cm dia gasoline-powered soil auger. The Shelby coring tube was then inserted in the cleared hole and driven approx. 12.7 cm into the ground to a

predetermined depth. Immediately after the removal of the Shelby tube from the hole, the clay in the bottom 7.6 cm of the core tube was transfered to an aluminum can 8.3 cm dia × 8.6 cm high. The can was then promptly sealed hermetically in the field with a commercial, hand-operated can sealer. Water immersion tests of sealed cans using this equipment at temperatures just below 100°C did not reveal any leaks. Time of collection and sealing were both noted. In most cases, a new hole was prepared for the collection of each sample. The collection process was repeated three or four times until either an obstruction was encountered or the maximum sampling depth used in this study (approx. 100 cm) was achieved. The samples were then transported as rapidly as possible (approx. 10–15 min) to the mobile laboratory where the γ-spectrometric counting equipment had been set up.

The prompt decay of the ^{214}Pb and ^{214}Bi in the canned samples was measured using shielded dual 7.6 × 7.6 cm NaI scintillation detectors and photomultipliers coupled through a multiplexer to a multichannel analyzer and printer. A detailed description of this dual NaI counting system has been published by Zelle *et al.*[6] The counting interval used was 1000 s unless otherwise noted and the measurements were taken on the ^{214}Bi 609 keV γ peak. At least three 1000 s counts were obtained on each of the samples in the first 2.5–3 h after sample collection. The prompt decay and subsequent build-up of the ^{214}Bi in the samples was followed by 10 or more additional measurements over a period of 600–700 h.

The ^{214}Bi counting data obtained on the samples are plotted on both linear and semi-logarithmic graph paper. In both plots the sum of the counts from the two NaI detectors corrected for background is plotted against the time that had elapsed from the collection of the sample to the midpoint of the specific 1000 s counting interval. The semi-logarithmic plot is used to estimate the ^{214}Bi counting rate at T_0, i.e. the counting rate of the 609 keV ^{214}Bi γ-peak in the sample for the 3- to 4-h period immediately preceding the collection of the sample.

The graphical estimate of the T_0 value is obtained either by the least-squares method or by fitting a straight line through the initial data points for the decay of the ^{214}Bi in each sample and noting the intercept of this line with the ordinate when the value of the elapsed time is zero. For some samples, particularly those which have experienced relatively small losses of ^{222}Rn during the sample collection and canning processes, this estimate at T_0 will closely approximate the actual ^{222}Rn/^{214}Bi concentration at T_0 in the sample. However, for those samples which have experienced significant ^{222}Rn sampling losses, the graphical T_0 estimate can be improved by noting the "y" coordinate of the intercept of the T_0 line and a vertical line with the general equation $x = a$ where "a" lies between 5 and 20 min. The selection of the value for "a" is directly related to the magnitude of the sampling loss. For a more detailed discussion of this point see the following section on the computer modeling of the prompt decay and build-up of the ^{226}Ra ^{222}Rn ^{214}Bi series.

It is important to emphasize here that in the prompt ^{214}Bi method it is assumed that in the 3- to 4-h period prior to the collection of the sample, the concentration of the ^{222}Rn has remained essentially constant. In this steady-state or quasi-equilibrium condition (not necessarily a condition of secular radioactive equilibrium) the ^{214}Bi counting rate and the number of atoms of ^{214}Pb and ^{214}Bi in the sample have also remained relatively constant. During this time period only the most recent additions of ^{222}Rn produced from the decay of the ^{226}Ra in the sample (a relatively small fraction of the total number of ^{222}Rn atoms in the sample) would not be in secular equilibrium with its short-lived daughter products, ^{214}Pb and ^{214}Bi. Further, during this short time interval, any small, recent additions of unsupported ^{222}Rn to the sample should be essentially balanced by corresponding losses of unsupported ^{222}Rn from the sample, leaving the ^{214}Bi counting rate attributable to this source essentially unchanged, i.e. the unsupported ^{222}Rn flux is essentially constant. The assumption also implies that if some of the *in situ* ^{222}Rn in the sample is being lost to the surroundings, the losses should be small and incremental rather than large and abrupt. This short-term, steady-state requirement of the prompt ^{214}Bi method does not preclude the long-term net loss or gain of ^{222}Rn from a soil sample, but rather it underscores the point that during this period small, incremental changes can be accommodated whereas large, abrupt changes in the *in situ* ^{222}Rn concentration, either losses or gains, will introduce uncertainties in the estimate of the *in situ* ^{222}Rn concentration.

In general, a large, abrupt loss of ^{222}Rn within the 3- to 4-h precollection interval cannot be distinguished, on the basis of the prompt ^{214}Bi measurements, from the sampling losses which may occur during the collection of the sample. The case of an abrupt pre-collection loss would yield a ^{214}Bi counting rate at T_0 somewhat greater than would be associated with the actual average *in situ*

^{222}Rn concentration in the sample for that period. The magnitude of this discrepancy would, of course, depend both on the net amount of ^{222}Rn lost and the time the loss occurred. On the basis of the prompt ^{214}Bi counting rate, the effects of such pre-sampling loss would be difficult to infer at $T_0 - 4$ h and would merge with the sampling losses as the pre-collection loss approached T_0. In practice, the assumption of an essential steady-state condition in the short time interval preceding the collection of the sample between ^{222}Rn and its daughters ^{214}Pb and ^{214}Bi seems to be supported both by the prompt ^{214}Bi measurements that have been made to date as well as the modeling studies.

COMPUTER MODELING OF THE PROMPT DECAY AND BUILD-UP OF THE ^{226}Ra/^{222}Rn/^{214}Pb/^{214}Bi SERIES

In order to understand in detail the interrelated processes of decay and build-up, a computer model of the decay and build-up of the part of the ^{238}U decay chain that contained the daughters ^{226}Ra, ^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po and ^{210}Pb has been developed. The model, based on the number of atoms of daughter products in equilibrium with 1 μg of ^{238}U, calculates for specified time intervals the (1) total number of decays of each daughter product accrued from T_0, (2) number of decays of each daughter product in the specified time interval, (3) number of atoms of each daughter product remaining at the end of a specific time interval and (4) total number of atoms of each daughter product present plus those formed during the time interval.

The model permits specification, at time T_0, of the initial, pre-collection deficiency or excess of ^{222}Rn when compared with the final ^{226}Ra/^{222}Rn/^{214}Bi equilibrium number of atoms or counting rate. The model also permits specifications of the sampling loss from 0 to 100% as a percentage of the initial number of atoms, at time T_0 originally present (or the counting rate). Finally, the model permits specification of the percentage of unsupported ^{222}Rn present and the percent sampling loss associated with this fraction of the ^{222}Rn in the sample at time T_0. This requirement is a consequence of the observation that, in general, the sampling loss associated with the unsupported ^{222}Rn fraction is almost always close to or equal to 100% whereas the sampling loss associated with the supported ^{222}Rn fraction is variable with a maximum loss at between 30–50% for most samples.

From Fig. 1 it can be seen that the simplest case does not require the model and occurs when the ^{226}Ra, ^{222}Rn and ^{214}Bi in a sample are in secular equilibrium and have experienced a zero sampling loss. In this case, the number of ^{222}Rn and ^{214}Bi atoms formed is equal to the number of ^{222}Rn and ^{214}Bi atoms decaying in any given time interval, and the ^{222}Rn/^{214}Bi counting rate at time T_0 is equal to the final ^{226}Ra/^{222}Rn/^{214}Bi counting rate. On either a regular or a semi-logarithmic plot of the counting rate or the number of atoms remaining vs time, the data points lie on a horizontal line passing through the number of atoms originally present at T_0 or the T_0 counting rate.

In addition, it can be seen in Fig. 1 that the semi-logarithmic plot for a sample that has experienced a steady-state, pre-collection ^{222}Rn loss of 25% and a ^{222}Rn sampling loss of 0%, also is nearly a straight line, does not pass through a minimum, and shows that the number of ^{214}Bi atoms for the first 1.5 h after T_0 remains essentially a constant, i.e. the line passing through the data points is essentially horizontal. Even in the first 10 h the increase in the counting rate or the number of atoms of ^{214}Bi for this case is essentially linear and has increased by only slightly more than 2%. It is this relationship that provides the basis for the statement that the contribution of ^{222}Rn from the decay of the ^{226}Ra in the sample in the 3–4 h preceding sample collection is relatively small.

Finally, for the case of the sample with a pre-collection ^{222}Rn loss of 0% and a 25% ^{222}Rn sampling loss, it can be seen in the semi-logarithmic plot (Fig. 1) that the prompt decay curve decreases at a relatively uniform rate between $T_0 + 900$ s and $T_0 + 4000$ s. Shortly after the minimum number of atoms (counting rate) has been reached, approx. $T_0 + 16{,}000$ s (Fig. 2), the 25% pre-collection loss and the 25% sampling loss plots coincide and begin a very slow, almost linear increase in the number of atoms (counting rate) for the next 5–10 h.

For a sample that has experienced a pre-collection loss, and has not lost ^{222}Rn during the sampling or canning process, the T_0 pre-collection number of ^{214}Bi atoms or counting rate for ^{214}Bi is obtained graphically by passing a straight line through the data points and noting the intercept on the "y" axis. Useful data for this graphical solution can be obtained as late as 5–10 h after T_0. This procedure yields a good T_0 estimate but is applicable to only a relatively small number of situations because most samples—even if they have experienced a pre-collection loss of ^{222}Rn—lose at least a little additional radon in the sampling process.

Figure 2 is a plot from T_0 to $T_0 + 40{,}000$ s of the prompt ^{214}Bi decay of samples in ^{226}Ra ^{222}Rn ^{214}Bi secular equilibrium which have experienced 10, 25, 50 and 100% losses of ^{222}Rn during the sampling process. The slopes of the linear segments of the curves passing through these sets of prompt ^{214}Bi data points (the T_0 lines or decay) are clearly a function of the percent sampling loss.

A detailed plot of the four different cases in Fig. 2 for the time interval T_0 to $T_0 + 7200$ s suggests that the optimum time to make the prompt ^{214}Bi measurement is from an elapsed time of approx. $T_0 + 900$ s (15 min) to an elapsed time of approx. $T_0 + 5400$ s (90 min). In this interval, the relationship between the data points for the full range of possible sampling losses is sufficiently linear to permit a good graphical estimate of the ^{214}Bi concentration or activity at T_0. The data from the model calculations for the interval T_0 to $T_0 + 1800$ s (30 min) suggest that, as the sampling losses increase, the estimate of the ^{222}Rn ^{214}Bi concentration at T_0 can be improved if the "y" intercept with the prompt ^{214}Bi decay line is obtained from a line parallel to the "y" axis with an equation of the general form $x = a$, where the constant "a" varies from approx. 300 to 1200 s, depending on the percentage of radon lost in the sampling process. For example, see Fig. 1. The offset is dictated by two factors: (1) the time required for the now unsupported ^{218}Po (half-life 3.05 min) to decay and (2) the percent sampling loss or the slope of the prompt ^{214}Bi decay line.

An estimate of the total sampling loss including both the supported and unsupported ^{222}Rn can be made based on the minimum ^{214}Bi counting rate which usually occurs approx. 4–4.5 h after sample collection (see Fig. 2). The sampling loss is a measure of the mobile, relatively weakly-held, supported ^{222}Rn in the sample and is obtained by subtracting the minimum ^{214}Bi counting rate (M) from the T_0 counting rate (T_0) and dividing by the T_0 counting rate, i.e. percent sampling loss = $(T_0 - M)\ T_0 \times 100$. This estimate includes the total amount of supported ^{222}Rn lost both prior to sample collection and during the sampling process.

Figure 3 shows the plot of the case where the calculated 25% sampling loss and 25% pre-collection steady-state loss have been combined. As in the example previously described, this plot of the combined losses after passing through its minimum, coincides with the plot for a sample that has

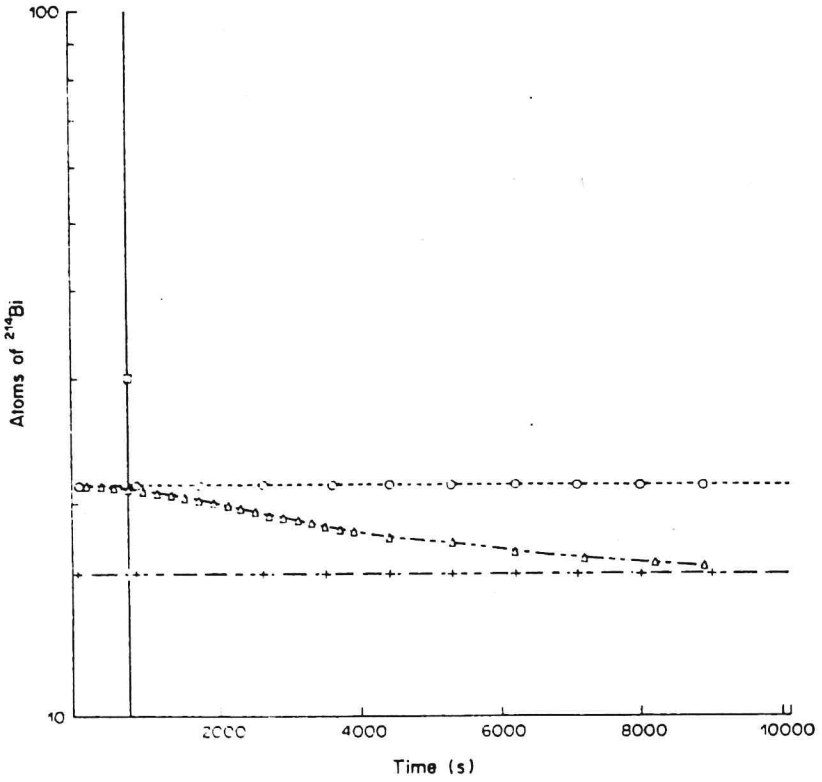

Fig. 1. Semi-logarithmic plot of the calculated number of atoms of ^{214}Bi remaining vs time for the following cases: ^{226}Ra ^{222}Rn ^{214}Bi equilibrium (○), a 25% sampling loss of ^{222}Rn (△) and a 25% pre-collection loss of ^{222}Rn with a 0% sampling loss of ^{222}Rn (+). The equation of the vertical line is $x = 800$ s (□).

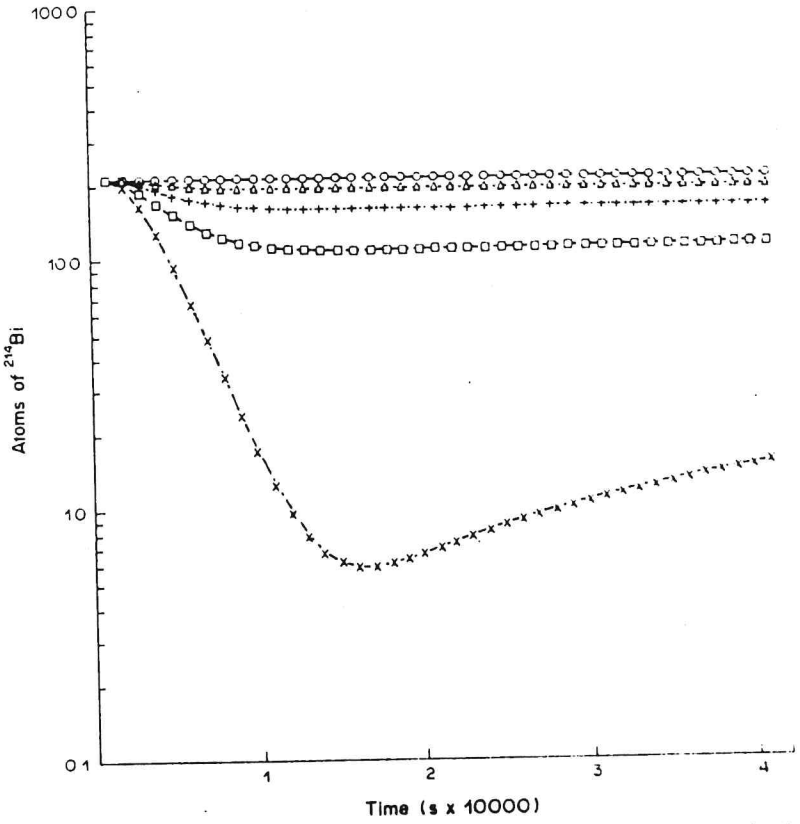

Fig. 2. Semi-logarithmic plot of the calculated number of atoms of ^{214}Bi remaining vs time for the following cases: 0% pre-collection loss of ^{222}Rn (○) and 10 (△), 25 (+), 50 (□) and 100(×)% sampling losses of ^{222}Rn.

experienced a pre-collection, steady-state loss of approx. 43.8%. An estimate of the ^{222}Rn deficiency in this sample at T_0 is obtained by subtracting the final equilibrium ^{214}Bi counting rate (Eq) from the ^{214}Bi counting rate at T_0 and dividing by the equilibrium counting rate, i.e. percent ^{222}Rn deficiency = $(Eq - T_0)/Eq \times 100$. An estimate of the mobile, weakly-held fraction of supported ^{222}Rn is obtained by subtracting the minimum ^{214}Bi counting rate (M) from the equilibrium counting rate (Eq) and dividing by the equilibrium counting rate, i.e. percent mobile fraction (supported) = $(Eq - M)/M \times 100$. This estimate approaches the emanating coefficient of the sample.

The most difficult case to evaluate quantitatively involves samples with overall pre-collection deficiency in ^{222}Rn which have received additions of unsupported ^{222}Rn. From the point of view of the prompt decay of ^{214}Bi, the unsupported ^{222}Rn which is in ^{222}Rn ^{214}Bi equilibrium is indistinguishable from the supported ^{222}Rn which is also in ^{222}Ra ^{214}Bi equilibrium. Any recently introduced unsupported ^{222}Rn, which has not yet had time to reach equilibrium with its ^{214}Bi daughter cannot, of course, be detected.

Samples which contain a pre-collection excess of unsupported ^{222}Rn are relatively easy to evaluate (see Fig. 4). An estimate of this excess, unsupported ^{222}Rn can be obtained by subtracting the final, equilibrium ^{226}Ra/^{222}Rn/^{214}Bi counting rate (Eq) from the counting rate at T_0 and dividing by the equilibrium counting rate, i.e. percent ^{222}Rn excess = $(T_0 - Eq)/Eq \times 100$. If, in this case, the sample experiences an additional sampling loss in which the minimum ^{214}Bi counting rate falls below the final equilibrium ^{214}Bi counting rate, the estimated excess of unsupported ^{222}Rn based on the equilibrium value will be the minimum estimate. The maximum estimate of the unsupported ^{222}Rn in the sample would be obtained by subtracting the minimum counting rate from the counting rate at T_0 and dividing by the counting rate at T_0, i.e. the total sampling loss would be assigned to the unsupported ^{222}Rn fraction and the pre-collection deficiency would be defined by the minimum counting rate.

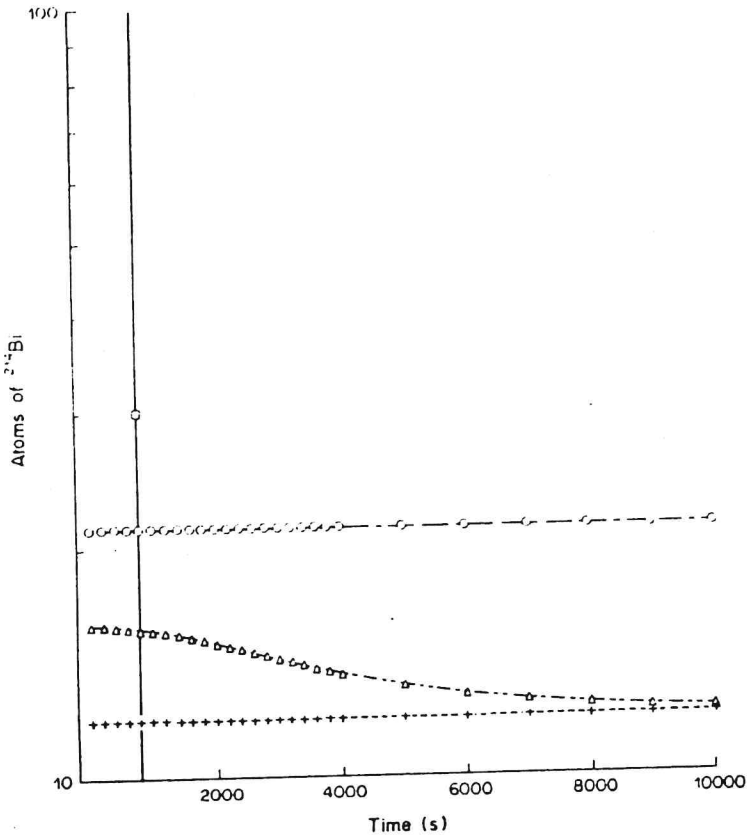

Fig. 3. Semi-logarithmic plot of the calculated number of atoms of ^{214}Bi remaining vs time for the following cases: ^{226}Ra/^{222}Rn/^{214}Bi equilibrium (○) a combined 25% pre-collection loss of ^{222}Rn (△) and a 25% sampling loss of ^{222}Rn (△) and a 43.75% pre-collection loss of ^{222}Rn with a 0% sampling loss of ^{222}Rn (+). The equation of the vertical line is $x = 1000$ s (□).

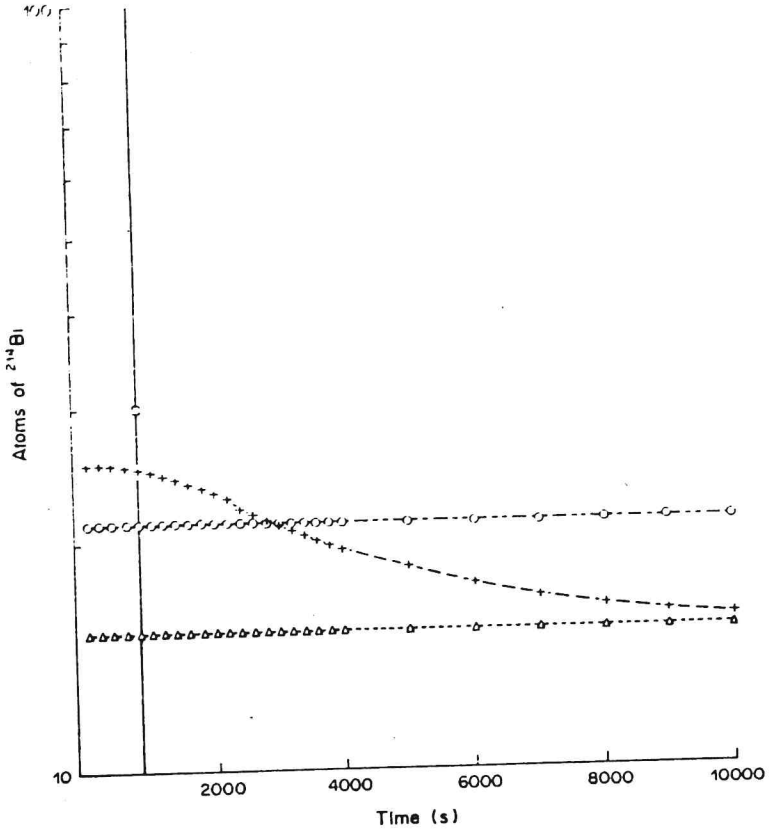

Fig. 4. Semi-logarithmic plot of the calculated number of atoms of ^{214}Bi remaining vs time for the following cases: ^{226}Ra/^{222}Rn/^{214}Bi equilibrium (○), a 20% excess of unsupported ^{222}Rn (+) and a 40% sampling loss of ^{222}Rn with a 0% sampling loss of ^{222}Rn (+). The equation of the vertical line is $x = 1000$ s (□).

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EARTHEN COVER TEST AREA MEASUREMENTS

Sample description and data

The 1981 earthen cover system was designed to test the effectiveness of four different ^{222}Rn barriers composed of 1.2 m thick layers of (1) Mancos Shale, (2) bentonite clay, (3) compacted adobe clay (each of these covered in turn with a 1.8 m thick layer of uncompacted adobe) and (4) a 3 m thick layer of uncompacted adobe clay. This test area is fully described by Hartley *et al.*[3] The samples for the prompt ^{214}Bi measurements were collected from four separate core holes in the 3 m thick cover of uncompacted adobe. The sample holes were located at the side of the column test facility access road that climbed the 3 m, uncompacted adobe cover at the southern end of the test area. The exact elevation of the cores above the tailings is difficult to establish but it is estimated that the collars of the core holes were at least 1.5–2 m above the tailings.

The semi-logarithmic plots of count rate vs time for the first 2 h 30 min of the measurements for samples EC-A, EC-B, EC-C and EC-D are given in Fig. 5. The linear plots of counting rate vs time up to 6 h for these four samples are given in Fig. 6. Figure 7 is a plot of counting rate vs time up to 700 h for samples EC-C and EC-D. The critical estimates of T_0, minimum and equilibrium ^{214}Bi counting rates are summarized in Table 1.

Discussion

The data from Table 1, and linear plots for samples EC-A, EC-B, EC-C and EC-D (Figs 6 and 7), are typical of samples that have large excesses of unsupported ^{222}Rn and show that, with the exception of sample EC-A, a very large fraction of the *in situ* ^{222}Rn in the samples is lost in the sample collection and canning process. The decay pattern of EC-A, the near surface sample (Fig. 6), shows a distinct minimum (3.12 count/g 10^3 s) before building-up to its final equilibrium value (3.88 count/g 10^3 s; Table 1). The equilibrium values for the remaining three samples, EC-B, EC-C

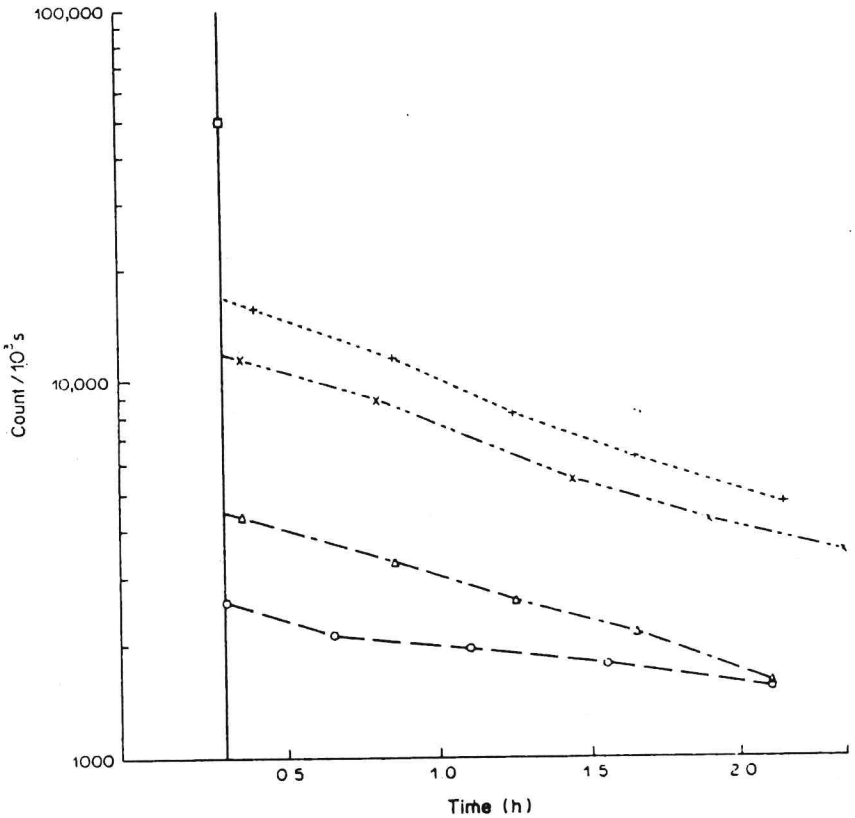

Fig. 5. Semi-logarithmic plot of the prompt ^{214}Bi count/10^3 s for samples EC-A (○), EC-B (△), EC-C (×) and EC-D (+) vs time from T_0, the time of collection, to T_0 + 2 h 30 min. The equation of the vertical line is x = 20 min (□).

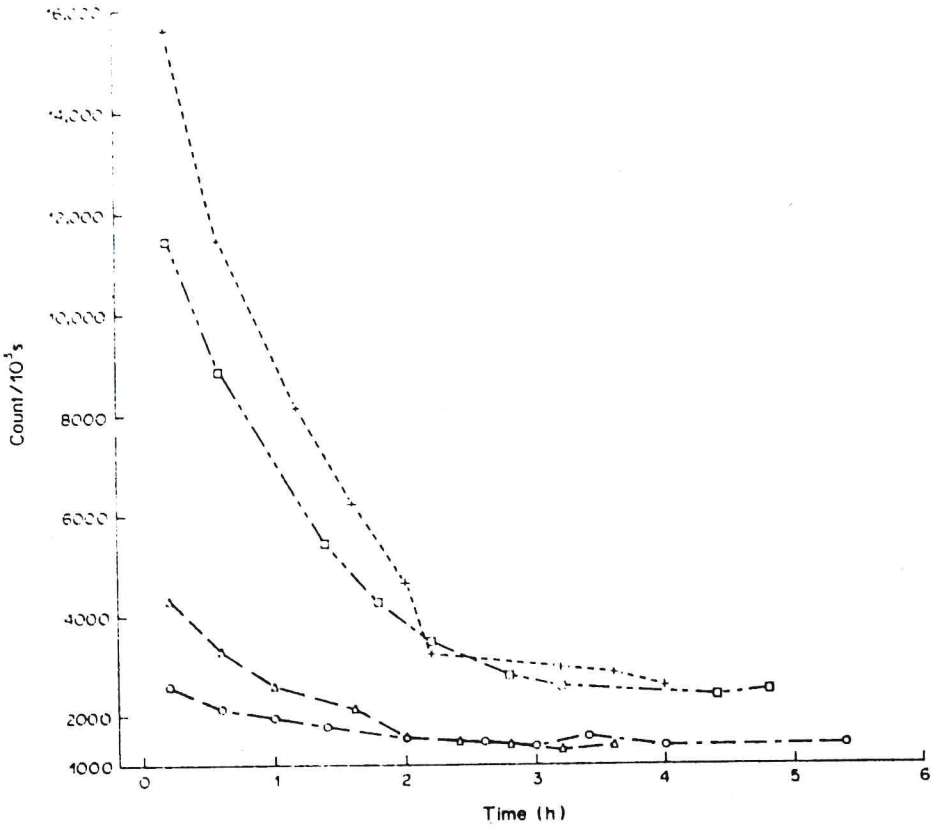

Fig. 6. Plot of the prompt ^{214}Bi count 10^3 s for samples EC-A (○), EC-B (△), EC-C (□) and EC-D (+) vs time from T_0 to $T_0 + 6$ h.

and EC-D [approx. 2.6–3.1 count g 10^3 s (see Table 1)] are thought to be representative of the average values for the original concentrations of ^{238}U and ^{226}Ra in the adobe clay. If this interpretation is accepted, then it would appear that EC-A has received some additional, probably unsupported ^{226}Ra, possibly as surface contamination. There does not appear to be any vertical increase in ^{226}Ra concentration with depth as might be expected if the underlying tailings were the source of the ^{226}Ra contamination.

In addition to experiencing a rapid initial decline in the ^{214}Bi counting rate as a result of major losses of unsupported ^{222}Rn in the sampling process, both samples EC-C and EC-D also exhibit a subsequent slow decay of the ^{214}Bi counting rates to their final equilibrium value [1454 and 1414 count 10^3 s, respectively (see Figs 6 and 7)]. This long-term decay pattern may be related to the presence of small amounts of residual unsupported ^{222}Rn which remained in the sample following the sample collection and canning process. The decay of the ^{214}Bi associated with this residual unsupported ^{222}Rn would, of course, be controlled by the 3.8 day half-life of ^{222}Rn.

The data on the prompt decay of the ^{214}Bi counting rate in these samples, as well as similar measurements made on other samples, lead to the conclusion that the unsupported ^{222}Rn in the samples is very weakly bound, is very mobile and is easily lost during the sample collection process. In this respect, the sampling losses associated with the unsupported ^{222}Rn are similar to the sampling losses associated with the interstitial, mobile, fraction of the supported ^{222}Rn resulting from the decay of the ^{226}Ra in the sample and deposited during the emanation process in the pore spaces of the sample.

The semi-logarithmic plots for samples EC-B, EC-C and EC-D (Fig. 5) are quite linear, yield good estimates of the *in situ* ^{222}Rn concentration at T_0, and are representative of samples in which the prompt ^{214}Bi decay is dominated by the unsupported ^{222}Rn lost in the sampling process. Only a relatively small contribution to the ^{214}Bi counting rate can be assigned to the original ^{226}Ra in the sample.

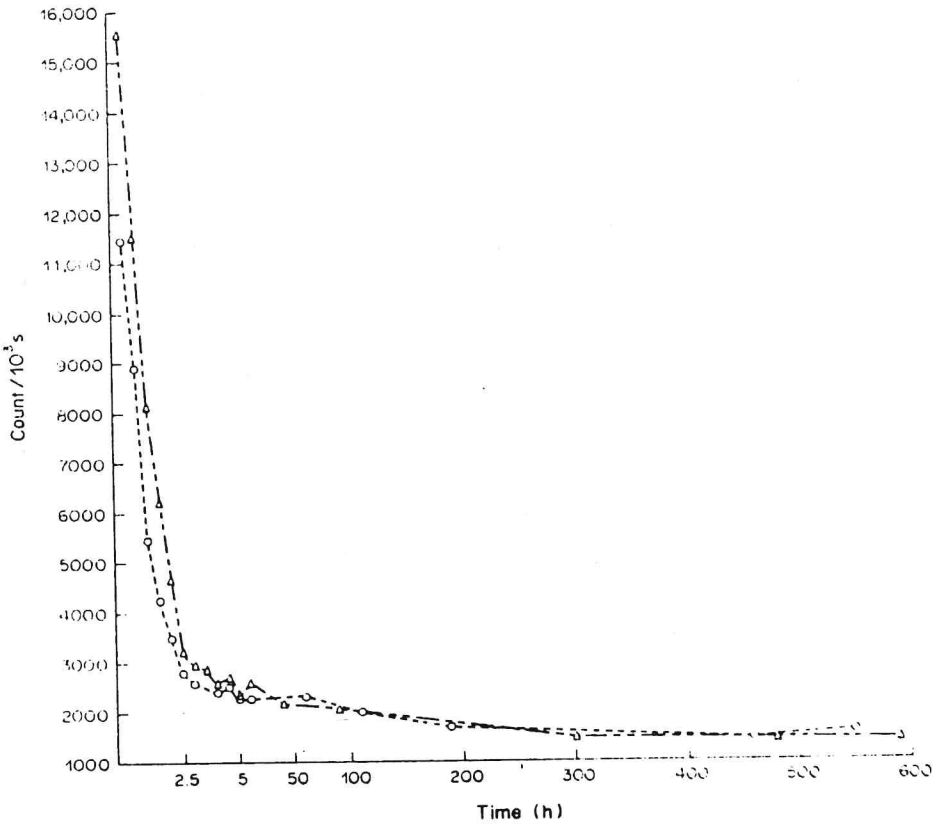

Fig. 7. Plot of the ^{214}Bi counts/10^3 s for samples EC-C (○) and EC-D (△) vs time from T_0 to T_0 + 700 h.

The data from the earthen cover test area is of special interest because it is now possible not only to make direct field measurements of the *in situ*, unsupported ^{222}Rn concentration in these samples but also to define, in a quantitative way, the changes in the concentration of unsupported ^{222}Rn as a function of sample depth. For example, Table 2 gives, for samples EC-A, EC-B, EC-C and EC-D, the excess, unsupported ^{222}Rn/^{214}Bi counting rates corrected for the amounts of supported ^{222}Rn in the samples at equilibrium. Table 2 also presents, based on the excess, unsupported ^{214}Bi counting rate,

Table 1. A summary of the T_0, minimum and equilibrium ^{214}Bi counting rates for samples FY-81 EC-A, EC-B, EC-C and EC-D from the earthen cover test area, Grand Junction tailings pile

Sample No. Depth (cm)	T_0 (counts/10^3 s) T_0 (count/g 10^3 s) Elapsed time (h) % ^{222}Rn pre-collection excess = $(T_0 - Eq)/Eq \times 10^2$	Minimum (count 10^3 s) Minimum (count g 10^3 s) Elapsed time (h) % ^{222}Rn sampling loss (total) = $(T_0 - M)\ T_0 \times 10^2$	Equilibrium (count 10^3 s) Equilibrium (count g 10^3 s) Elapsed time (h) % ^{222}Rn mobile fraction (supported) = $(Eq - M)\ Eq \times 10^2$
EC-A 5.1–12.7	2680 (x = 15 min) (6.06) 00:00:00 56.4% (excess)	1378 (3.12) 03:10:05 48.6%	1714 (3.88) 561:36:25 19.6%
EC-B 30.5–38.1	4570 (x = 20 min) (9.85) 00:00:00 220% (excess)	1307 (2.82) 03:22:50 71.4%	1429 (3.08) 664:00:50 8.53%
EC-C 55.9–63.5	12,000 (x = 20 min) (21.58) 00:00:00 725% (excess)	Two small intermediate minimums — — —	1454 (2.61) 682:38:15 —
EC-D 94.0–101.6	17,500 (x = 20 min) (35.25) 00:00:00 1140% (excess)	2586 Two small intermediate minimums — —	1414 (2.85) 596:39:40 —

Eq = Equilibrium ^{214}Bi counting rate. T_0 = T_0 ^{214}Bi counting rate. M = Minimum ^{214}Bi counting rate. x = 15 min and x = 20 min equations of vertical lines used to obtain T_0 counting rate.

Table 2. A comparison of the excess, unsupported T_0 ^{222}Rn ^{214}Bi (count g 10^3 s) and the inferred ^{222}Rn concentration in pCi cm^3 in samples EC-A, EC-B, EC-C and EC-D with the calculated concentration in pCi cm^3 of ^{222}Rn obtained from the RACOM radon diffusion model

Sample No Depth (cm)	T_0 excess ^{222}Rn ^{214}Bi (observed) (count g 10^3 s)	T_0 concentration of ^{222}Rn (inferred) (pCi/cm^3)	Concentration of ^{222}Rn (calculated) (pCi/cm^3)
EC-A	2.18	7.7	7.0
EC-B	6.**	23.8	27.6
EC-C	18.9*	66.8	50.1
EC-D	32.4	114.1	90.7

* Excess ^{222}Rn ^{214}Bi = T_0 (count g 10^3 s) − F_4 (count g 10^3 s) (see Table 1).

the inferred concentrations of the excess, unsupported ^{222}Rn (pCi/cm^3) in the total pore space of the samples (assumed clay porosity = 0.40). Table 2 also gives, for comparative purposes, the calculated average concentration in the four samples of the pCi/cm^3 of ^{222}Rn in the total pore space using the computer code RAECOM.[7] The RAECOM calculations assume a clay porosity of 0.4, a 10% moisture content, a density for the clay of 1.62 g/cm^3, an emanation fraction of 0.30, and an assumed ^{226}Ra concentration in the underlying tailings of approx. 1000 pCi/g.

From Table 2 it can be seen that the agreement between the initial calculations of the ^{222}Rn concentrations in pCi cm^3 from the diffusion model and the pCi/cm^3 of ^{222}Rn based on the T_0 excess ^{222}Rn ^{214}Bi counting rates is reasonably good. It would appear from this agreement that the initial model assumptions were not unreasonable. Reductions in the discrepancies between the two sets of values, particularly for samples EC-C and EC-D, could be achieved by a number of different adjustments in the model assumptions including increasing the tailings ^{226}Ra concentration from

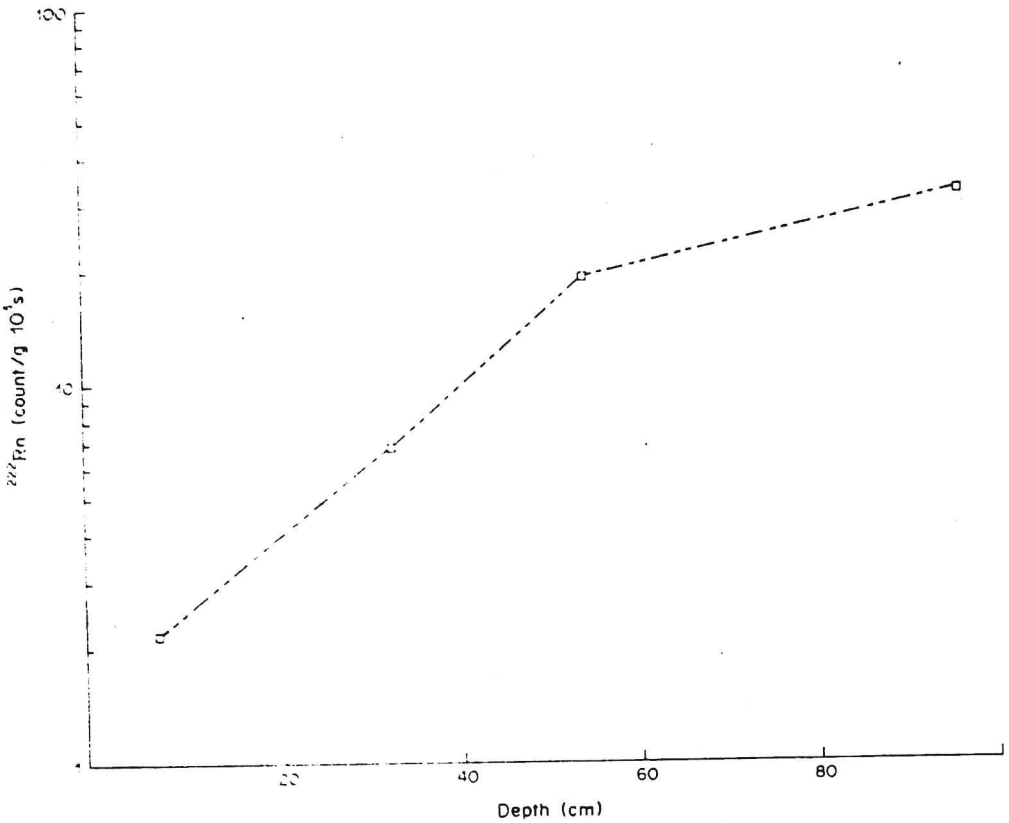

Fig. 8. Plot of the excess, unsupported ^{214}Bi counts g 10 s for samples EC-A, EC-B, EC-C and EC-D vs depth of the sample (cm).

1000 to 1200 pCi/g or increasing the emanating fraction from 0.30 to 0.35. Both changes would fall well within the observed range of these values at the Grand Junction tailings pile. If, however, the agreement between the two sets of observations is improved for samples EC-A, EC-C and EC-D, it appears that the disagreement between the two sets of values for EC-B will increase.

Finally, the RAECOM model gives a calculated ^{222}Rn flux out of the surface of approx. 63 pCi m^{-2}. This value may be compared to an average value of approx. 75 pCi m^{-2} s obtained from long-term flux measurements made over the uncompacted adobe cover. The example noted above suggests that if the measured and calculated profiles of unsupported ^{222}Rn concentration are in reasonably close agreement, the calculated ^{222}Rn flux from the surface based on the prompt ^{214}Bi measurements, should also be in general agreement with traditional surface flux measurements.

SUMMARY AND CONCLUSIONS

It is clear that in order to more fully evaluate the potential of the prompt ^{214}Bi technique, many additional field and laboratory tests must be undertaken. However, the available data from this preliminary set of measurements suggest the following:

1. A good, indirect measurement of the *in situ* concentration of ^{222}Rn at the time of sample collection, T_0, can be made based on a field method of measuring the prompt decay of ^{214}Bi counting rate in sealed samples of soil or tailings.
2. A good, indirect measurement of the state of ^{226}Ra-^{222}Rn equilibrium at the time of sample collection can be made based on the T_0 measurement of the ^{222}Rn-^{214}Bi concentration and subsequent ^{214}Bi measurements made after the ^{226}Ra in the sealed samples of soil or tailings has re-established secular equilibrium with the ^{222}Rn and ^{214}Bi in the samples.
3. A good estimate of the concentration of excess, unsupported ^{222}Rn in samples of soil or tailings at the time of sample collection (a special case of Conclusion 2 above) can be made based on the difference between the T_0 and the final, equilibrium ^{214}Bi counting rates.
4. Estimates of the ^{222}Rn lost in the sampling process and the fraction of mobile, relatively weakly held, supported ^{222}Rn in samples of soil or tailings at the time of sample collection can be made based on a measurement of the prompt minimum ^{214}Bi counting rate and either the T_0 or the final ^{226}Ra/^{214}Bi equilibrium counting rate.

Should additional field and laboratory tests of the prompt ^{214}Bi technique support the preliminary measurements that have been made, it should be possible, using the data from the T_0, minimum, and final equilibrium ^{214}Bi counting rates, to do the following:

(i) Provide a means of directly measuring in the field the concentration of both supported and excess, unsupported ^{222}Rn in soils or tailings samples as a function of the depth of the sample and of calculating the surface ^{222}Rn flux based on the gradient data.

(ii) Provide a field method capable of studying the ^{222}Rn transport mechanisms in soils and uranium mill tailings as well as a laboratory method capable of measuring experimental, unsupported ^{222}Rn concentration gradients and studying the diffusion and adjective components of radon transport.

(iii) Determine the effectiveness of specific tailings cover designs as ^{222}Rn barriers on the basis of measured excess, unsupported ^{222}Rn concentration gradients and the calculated ^{222}Rn flux from the surface of the cover and provide an additional tool for use in monitoring the long-term performance of tailings covers used in the U.S. Department of Energy Uranium Mill Tailings Remedial Action UMTRA program.

(iv) Provide a field method for measuring the concentrations of ^{226}Ra and *in situ* ^{222}Rn in soil profiles at building sites prior to the start of construction, characterizing the mobility of the ^{226}Ra and *in situ* ^{222}Rn in these profiles.

Provide a means of directly measuring in the field the concentration of b

calculating the maximum and minimum ^{222}Rn surface flux and providing an improved basis for assessing the potential hazard from the mobile, unsupported ^{222}Rn at such sites.

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