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Authors

Nelson, P.H.

Rachiele, R.

Smith, A.

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TRANSPORT OF RADON IN FLOWING BOREHOLES AT
STRIPA, SWEDEN

P. H. Nelson, R. Rachiele, and A. Smith

Earth Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

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ABSTRACT

Granitic rock in an underground experimental waste storage site at Stripa, Sweden is unusually high in natural radioelements (~40 ppm uranium) with higher concentrations occurring locally in thin chloritic zones and fractures. Groundwater seeping through fractures into open boreholes is consequently highly anomalous in its radon content, with activity as high as one microcurie per liter. When total count gamma-ray logs are run in boreholes where groundwater inflow is appreciable, the result is quite unusual: the radon daughter activity in the water adds considerably to the gamma contribution from the rock, and in fact often dominates the log.

The total gamma activity increases where radon-charged groundwater enters a borehole, and decays as the water flows along the hole in response to the hydraulic gradient. As a consequence, the gamma log serves as a flow profile, locating zones of water entry (or loss) by an increase (or decrease) in the total gamma activity. If mixing within the borehole does not occur, the activity decreases exponentially along the hole away from the entry point because of the steady decay of radon and its daughter products as they migrate with the flow in the water column. This spatial decay rate can be converted to a linear flow rate since the 3.8-day half-life of radon governs the response time. For example, if the volumetric flow rate in a 76-mm hole falls within the range 0.5 to 50 liters per day, and if observations are available from a 10 m length of hole, then the flow rate can be measured quantitatively. Proportionately higher rates can be measured if longer hole lengths are available for observation.

A model for flow through a thin crack emanating radon at a rate E shows that the radon concentration of water entering a hole is $E/\lambda h$, where λ is the radon

decay rate and h the crack aperture, assuming that the flow rate and crack source area are such that an element of water resides within the source area for several radon half-lives or more. Using this simple relationship, independent measurements of emanation and concentration produce reasonable estimates of fracture aperture. Although uranium concentration values at Stripa are unusually high, neither the emanation coefficients nor the fracture properties appear to be unusual for granitic rock. It therefore seems likely that many granitic sites must exist where the radon content in groundwater is higher than in other geological terranes, although perhaps not as high as the microcurie per liter concentrations found at the Stripa site.

INTRODUCTION

Our findings on radon migration in groundwater came about during the course of borehole logging investigations at an experimental site for waste storage investigations. The site at Stripa, located in central Sweden, was until recently an active iron mine, but is now dedicated to hydrological and rock mechanics investigations for the disposal of radioactive waste (Witherspoon, Cook and Gale, 1980). For hydrological and in situ stress measurements, four cored holes were drilled from surface, at angles ranging from 44 to 90 degrees from horizontal, to slant depths ranging from 320 to 380 meters (Figure 1). For the underground experiments, over 100 boreholes with lengths ranging from 10 to 30 meters were drilled in three drifts located at a depth of 340 meters. Prior to installation of the fixed experiments during 1978, gamma-ray and other geophysical logs were acquired in many of these boreholes (Nelson, et al, 1979).

The boreholes drilled from surface penetrate leptite, a metamorphosed sedimentary and volcanic sequence, and the quartz monzonite of the Stripa pluton. The underground experimental drifts are located completely in quartz monzonite, about 50 to 100 meters from the leptite-quartz monzonite contact which dips to the southeast. The experimental drifts are approached from a drift in the now inactive iron mine. The iron ore, which has been extensively mined, is stratiformly bound within the leptite which forms a folded and faulted synclinal structure with a gently plunging ENE axis (Olkiewicz, 1979).

Initially the objective of the borehole logging program, combined with visual observation of core, was to provide physical measurements which could be used to infer the mechanical and hydrological character of the site. The

gamma-ray probe was one of several logging probes applied for this general purpose. However, the presence of several inexplicable features in the gamma-ray logs, the findings of high radon content in groundwater (Fritz, et al, 1979), and the unusual distribution of radioelements in the Stripa quartz monzonite (Wollenberg, et al, 1982) caused us to pursue more carefully the impact of anomalous radon content in the Stripa groundwaters upon the borehole gamma-ray records (Nelson, et al 1981).

Radon is the only gas in the uranium and thorium decay series and is not normally ionic in water; consequently it is more mobile than other members of the series and is often responsible for disequilibrium conditions. Because of its uniqueness as the only naturally-occurring radioactive gas present in the earth's crust, it has been singled out for study as a remote indicator of economic concentrations of uranium (Fleisher and Magro-Campero, 1978) and as a possible precursor of earthquake activity (King, 1980). The mechanisms of radon emanations and subsequent migration in rock have been reviewed by Tanner (1964; 1978). However, a fully satisfactory explanation of its transport through rock remains a subject open to further investigation (Kristiansson and Malmqvist, 1980).

Some fraction of the radon emanated from rocks will migrate into systems of connected pores or fractures, and hence enter the groundwater regime. This radon may then be transported for tens or even hundreds of meters from its birthplace before decay (3.82 day half-life), to appear at concentrations which may or may not reflect the radon emanation of local aquifer materials. Groundwater radon concentrations ordinarily span at least the range 10 to 10,000 pCi/liter; both lower and higher values are found, but are relatively rare. In

terms of country rock lithology, the lowest values are found in ultramafic terranes, and excluding zones of uranium mineralization the highest values are found in acidic granitic terranes. In other words, groundwater radon concentration is roughly in proportion to the uranium content of the source rock. However, special circumstances such as severe chemical weathering, hydrothermal solution deposition, or extensive fracturing can produce dramatic increases in groundwater radon concentrations above those expected from the bulk rock uranium content.

RADIOELEMENT DISTRIBUTION IN ROCK

Measurements of uranium, thorium, and potassium concentrations in core samples were performed at the LBL Low Background Facility, using a high-sensitivity gamma spectrometer to analyze samples of 800-1000 gram weight. In this system, events detected in an 8-in diameter by 4-in thick NaI(Tl) crystal are processed by a gain-stabilized multichannel pulse height analyzer to produce 400-channel spectra that span the gamma energy range from 0.1 to 4.1 MeV.

A gamma spectrum of a sample of crushed core from Stripa is shown in Figure 2. Selected intervals from such spectra (indicated on Figure 2) are processed by a set of linear equations to yield the radioelement content, where the absolute calibrations are based on equilibrium ores certified by the New Brunswick Laboratory (USDOE) for uranium and thorium, and on CP grade potassium chloride for potassium.

The gamma spectrum emitted by the rock is extremely complex, containing hundreds of spectral lines (Smith and Wollenberg, 1971). This complexity is obscured in Figure 2, however, because most of the individual lines are

of such low intensity that only a few dominate the observed spectrum. In addition, the intrinsic characteristics of NaI scintillation crystals severely limit the obtainable resolution.

Average value and ranges of spectral assays given in Table 1 were obtained from spectra such as that in Figure 2 using the indicated gamma energy intervals and applying spectrum stripping techniques. Fifty-eight core samples 10 cm in length were cut from core obtained from borehole N1, a 12.8 m hole drilled vertically from one of the underground drifts. Three samples, dark in color and high in chlorite, gave values considerably higher than the other 55 and are averaged separately. Average U, Th, and K values of the remaining 55 were 38.7 ppm, 26.9 ppm and 4.03% with respective standard deviations of 8.9 ppm, 7.4 ppm and 0.41%. These amounts of U and Th are much higher than in most granitic rocks: typically the concentration of uranium is in the 3 to 15 ppm range and the Th/U ratio ranges from 3 to 5.

Also shown in Table 1 are data from 24 crushed samples, including one check sample from borehole N1. The samples were processed in a jaw crusher to reduce the size of the largest fragments to about -1/2 inch; all finer sizes were retained in the samples. The samples from underground holes V-1 and N1 and surface borehole SBH-1 are quite comparable in U, Th, and K values to the whole core results. Samples from SBH-2 have about one-half the U content, but these samples were selected from short zones where the total count gamma-ray borehole log registered a low count rate. Except for these few occurrences, the core samples and the borehole logs indicated that the uranium concentration is quite uniform throughout the drilled portion of the Stripa quartz monzonite.

Because the γ -rays used for U-assay originate from decay of Rn-222 daughter nuclides, two analyses of a single sample can also provide a measure of emanating Rn-222: the first, while Rn-222 is free to emanate; the second, at a suitable time after sealing against Rn-222 escape. The difference between the two apparent U-concentrations can be converted into a value for Rn-222 emanation. The data are given in Table 1 as the fractional value lost from the sample and as activity per gram of rock. Although for very small particles the emanation rate of Rn-222 is known to be sensitive to particle size, no significant fraction of these materials is believed to be in the size range for which such effects are important. The observed Rn-222 is more likely to be related either to the degree of chemical weathering, or to radiation damage that has accrued over the formation's long life, due to decay of the unusually high concentration of uranium.

The in situ gamma-ray activity was measured with a total count borehole probe manufactured by Mt. Sopris Instrument Co. of Delta, Colorado. Spectral capability would have been preferred but no spectral probe was available within the constraints of the field project. The gamma probe is 32 mm in diameter and 2.07 m in length. Gamma rays are detected by a 12 mm x 38 mm NaI(Tl) scintillation crystal. The detector circuit has no adjustable discriminator setting; the low energy cut off is estimated to be about 50 keV. Pulses produced by the probe were counted and integrated by a rate-meter and the output recorded in the form of counts per second on a chart recorder. Logs were recorded at a depth scale of 50:1 or 1/2 m of borehole per 1 cm of chart paper. Averaging time was 4 sec at a logging speed of 2.5 m/min. Logs were acquired at a scale of 50 counts per second (cps) per cm of chart paper with a backup log at a scale of 25 cps per cm. Dead-time compensation circuitry in the ratemeter was not needed and was not used.

Pulses could also be counted for a specified period of time by a Canberra model 1775 nuclear counter operated in parallel with the ratemeter.

Gamma-ray logs from 18 vertical boreholes in one of the underground drifts are shown in Figure 3. All boreholes were water-filled when logged. Boreholes prefixed with a U or T are 38 mm diameter, with an E, 76 mm; and with an H, 406 mm. All the gamma ray baselines in this figure are given as 200 cps, with the exception of heater hole H9 where the count was much reduced due to the increased attenuation in the water-filled, large-diameter (406 mm) hole. Most logs are within 20 cps of the baseline, demonstrating the uniformity of the U and Th distribution mentioned previously. Borehole E8 registers a count rate above baseline which we attribute to the presence of dissolved radon. The horizontal and vertical spatial scales are the same in Fig. 3, thereby portraying the logs superposed on a true cross-sectional view of the rock mass, except for the break in the cross-section between holes E8 and E13. One prominent geologic feature can be seen. An anomalous decrease in the count rate with a half-width of less than 0.5 m reveals a planar feature dipping downwards from right to left in holes E6 through U2. This feature is caused by a pegmatite dike, which in most granitic rock would produce a count rate increase due to the higher potassium content of pegmatites. Here, however, the count rate in the pegmatite decreases because its uranium and thorium content is lower than that of the quartz monzonite.

RADIOACTIVITY IN GROUNDWATER

Unlike the majority of records shown in Figure 3, other gamma-ray borehole logs from Stripa are well above the 200 cps rate for much of their length.

Most notable are several logs from a second experimental drift located about 20m from the first. During the heater experiment, water was regularly removed from the heater holes. It was observed that those with high overall gamma count rates were also the holes with high water inflow rates. Figure 4 demonstrates a good linear correlation between water inflow rate and gamma count rate. Thus the gamma log was recognized as a potential water flow indicator at Stripa.

Radon dissolved in water was the suspected cause of the exceptionally high count rates. To confirm that radon was being transported by water into the boreholes, a sequence of logs was run in borehole M3, a 38 mm diameter hole located at the rear of the second drift. Borehole M3 was unusual in that it had an exceptionally high flow rate among the underground boreholes: measurements of its flow rate ranged from 124 to 216 liters per day during the two-year observation period. The water was removed from M3 and several gamma ray logs were run over a period of a day as water infiltrated back into the hole, causing the water column to rise. A gamma log recorded prior to dewatering and the logs run sequentially after the dewatering are shown in Fig. 5. Low count rates were observed with the probe in air, high rates in water, and a shelf reflecting the air-water interface can be seen rising upwards progressively in the three logs obtained 0:06, 1:02, and 1:33 (hours: minutes) after water removal. The rising shelf confirms that water flowing into the holes was responsible for the high gamma activity seen in the logs.

To identify radon as the specific radioelement being transported by water, a water sample from M3 was collected in a 60-liter vessel of 0.18 m radius and the gamma-ray probe inserted into it through a rubber stopper. The count rate was then monitored for 13 days as shown in Fig. 6. The slope of the straight line is determined by the decay rate of radon-222.

Radon analyses of M3 water have been reported by Fritz et al (1979). Table 2 shows that the M3 sample yielded radon-222 values of 1.9 and 1.3 microcuries per liter from analyses performed in two laboratories. The concentration level is extremely high, some three to six orders of magnitude greater than radon concentrations normally found in groundwater. These unusually high concentrations help explain why there are almost no similar observations reported in the literature. The field study by Løvberg et al. (1980) is the only other study known to us where radon in the borehole was reported to perturb the gamma ray measurement.

Although the 3.8-day radon half-life controls the time-dependent aspects of the gamma-ray measurement, it is the daughters of radon-222, bismuth-214 and lead-214, which produce the observed gamma radiation. In a flowing borehole these products could become spatially segregated from their radon parent, thereby complicating the decay patterns. However at the low flow rates observed in this study, such segregation, if it exists, is not very significant. There is also the possibility of cations plating onto the probe itself. This was not observed, however, as bi-directional passes of the probe through high count zones did not appear to disturb the patterns.

MODELS OF RADON TRANSPORT IN FLOWING BOREHOLES

The radon concentration observed in flowing boreholes will be controlled by two factors, the concentration of the radon in water at the time it enters the holes, and the mixing that occurs after entry. The entering concentration will be determined by the emanating power of the rock, the quantity of water moving through the emanating region, and the length of time which a given volume of water spends within the emanating region. As our data were gathered in igneous rocks where flow is along fractures, we first develop

a simple model of flow along a single planar fracture containing a source of radon. Because flow is the only hydrological quantity measured, the model assumes that only flow is known; reservoir and wellbore pressures are not included.

Radial Flow Along a Thin Crack

A simple crack model (Fig. 7) can be used to better understand the relation between flow rate and radon concentration. We follow the development of Stoker and Kruger (1975) for porous media, adapted to a thin, flat disk of aperture h which intersects a vertical hole of radius a . Water flows radially inward from all directions through the crack to be collected within the central hole. As it flows inward, it picks up a fraction of the radon atoms produced by radium present on the surfaces of the fracture. The rate of radon production is called the emanating power, expressed as the effective radium (not radon) activity per unit area, or equivalently, as the production of radon per unit time per unit area.

The parameters employed in the crack model are:

h	crack width (cm)
a	borehole radius (cm)
A	cross-sectional area of borehole (cm^2)
b	disc radius (cm)
r	radial distance from borehole axis (cm)
z	axial distance along borehole (cm)
Q	volumetric inflow (cm^3/day)
E	emanating power (number of radon atoms produced per cm^2 per day)
λ	decay constant of radon-222 (0.18/day)
I	radon influx (radon atoms/day)
C	radon concentration (radon atoms/ cm^3)
C_0	radon concentration of water entering borehole

If an elemental annulus of width dr (Fig. 7) produces radon at a rate E , then its contribution to the influx of radon atoms entering the borehole will be:

$$dI = Ee^{-\lambda t} 2\pi r dr \quad (7)$$

where the time for fluid to move from the annulus to the borehole is:

$$t = \frac{1}{-Q} \int_r^a 2\pi h r dr = \frac{\pi h}{Q} (r^2 - a^2) \quad (8)$$

If radon is produced from a disc source extending outwards from the borehole, then the influx I is obtained by integrating the above expression over its inner radius a to its outer radius b . The result is:

$$I = \frac{QE}{\lambda h} \left(1 - e^{-\lambda \pi h (b^2 - a^2)/Q} \right) \quad (9)$$

and the concentration C_o in the borehole is simply I/Q ,

$$C_o = \frac{E}{\lambda h} \left(1 - e^{-\lambda \pi h (b^2 - a^2)/Q} \right) \quad (10)$$

The flow rate and the source geometry determine the value of the exponent and hence the dependence of the concentration upon flow. If the flow is sufficiently high so that

$$Q \gg \lambda \pi h (b^2 - a^2) \quad (11)$$

then,

$$C_o \approx E \pi (b^2 - a^2) / Q \quad (12)$$

and the concentration increases linearly with the source area and decreases inversely with Q . This condition occurs if the source area is small enough so that the fluid crosses it in a fraction of the radon half-life. The $1/Q$ dependence is quite contrary to our observations at Stripa, hence the high flow condition does not apply to conditions at the Stripa site.

On the other hand, if the flow rate is low enough or the source area large enough that the fluid residence time within the source area is several half-lives or more, so that

$$Q \ll \lambda \pi h (b^2 - a^2) , \quad (13)$$

then the exponent can be ignored and

$$C_o = E/\lambda h . \quad (14)$$

Hence, the concentration is independent of flow rate and dependent upon geometry only through the fracture width h . Assuming a flow rate Q of 10 liters per day and a value of h of 0.01 cm, the low flow approximation requires that the outer radius of the disk be at least 13 m. Because there is good field evidence that the radioelement concentration is more or less uniform over a 100 m scale, it is apparent that the low flow approximation applies to the data under consideration here. Therefore, we expect the radon concentration of groundwater entering each borehole to be reasonably constant, subject only to local variations in radon emanation and crack aperture.

Once radon-charged groundwater has entered the borehole, its distribution within the borehole will depend in a complicated way upon the points of entry, the flow rate, the diffusion of radon in water, and dispersion occurring during flow. It is likely in some situations that convective overturn within the hole

will also further mixing. We ignore diffusion and dispersion in developing a few simple models for comparison with the field data. The first case considered is the intersection of the borehole by a single fracture, with either uniform, non-dispersive flow or complete mixing within the borehole. This case is readily extended to examine complete mixing within the borehole. And thirdly, we examine the case of continuous water entry along the length of the borehole.

Borehole Concentration For a Single Entry Point, No Mixing

If radon enters at one depth within a borehole and moves upward within the water column without diffusion or dispersion, then a simple exponential decay with distance z from the source should be observed. The travel time from the entry point to the observation point in a hole of area A is simply Az/Q , so the concentration is

$$C(z) = C_o e^{-\lambda Az/Q} \quad (15)$$

or

$$\ln [C(z)/C_o] = -\lambda Az/Q \quad (16)$$

Since gamma-ray count is proportional to concentration $C(z)$, Eq. (16) suggests that the logarithm of the gamma-ray count rate be plotted against depth. Then a straight-line fit to the decay will give the flow rate Q if the single-entry, no-mixing conditions prevail. For example, if z represents the distance over which the count rate decreases by half, then Eq. (16) shows that Q is $\lambda Az/0.693$.

Figure 8 shows Eq. (16) plotted for five linear flow velocities, Q/A . The volumetric flow rate Q is also given for a 76-mm diameter hole. Along a 14-m travel path, different flow rates can be distinguished only within the range 0.1 to 10 m/day. Above 10 m/day there is less than 20% drop over a 10 m length; at higher velocities we can determine only that the flow rate is greater than 10 m/day. At rates below 0.1 m/day dispersion, sensitivity and water displacement by the probe will limit the detection of anomalous concentrations. Figure 8 applies to longer or shorter travel paths simply by scaling: over a 140 m path we could evaluate flows ranging between 1 and 100 m/day.

Complete Mixing

If the inflow Q from a single entry point, or from any distribution of multiple entry points, is somehow mixed throughout a volume V , then the observed concentration will be an average of all contributions, allowing for the amount of decay during the mixing time. This model encompasses the case where the entry point is a fracture at the bottom of an artesian borehole, with complete mixing occurring before an entering elemental volume of water exits at the collar.

For a mixing time of T , the average concentration will be:

$$\bar{C} = \frac{1}{T} \int_0^T C_0 e^{-\lambda t} dt \quad (17)$$

or,

$$\bar{C} = \frac{C_o}{\lambda T} (1 - e^{-\lambda T}) \quad (18)$$

The mixing time can be no less than the time required to fill the volume, V/Q , in which case,

$$\bar{C} = C_o \frac{Q}{\lambda V} (1 - e^{-\lambda V/Q}) \quad (19)$$

This result is shown graphically in Fig. 9, and is discussed in conjunction with the model for continuous uniform entry in the next subsection. Values of λV are given in Table 3.

Continuous Uniform Entry

Assume that water enters the borehole uniformly along its entire length rather than at a discrete entry point. At the time of entry, each elemental volume of water bears radon at a concentration C_o . Since the infiltration rate is q (l/m-day), the volumetric flow along the axis of the hole is $Q(z) = qz$ and the linear velocity is $v(z) = qz/A$. The water originating from an elemental length dz' located at z' will occupy a fraction dz'/z of the volume passing an observation point z . The time required to transport the elemental volume from the entry point z' to z is:

$$t' = \int_{z'}^z \frac{dx}{v(x)}$$

or:

$$t' = \frac{A}{q} \ln(z/z') \quad (20)$$

The concentration at z due to the contribution from z' is

$$\begin{aligned} C' &= C_o e^{-\lambda t'} \\ &= C_o (z'/z)^{\lambda A/q} \end{aligned} \quad (21)$$

Integrating over all contributions between 0 and z ,

$$\begin{aligned} C(z) &= \frac{1}{z} \int_0^z C_o \left(\frac{z'}{z}\right)^{\frac{\lambda A}{q}} dz' \\ &= C_o \frac{q}{q + \lambda A} \end{aligned} \quad (22)$$

Hence the concentration is constant along the borehole length, for any specified q and A (or equivalently, Q and V). In terms of total flow, Eq.

(22) can be written:

$$C(z) = C_o \frac{Q/\lambda V}{1 + Q/\lambda V}, \quad (23)$$

which is compared with Eq. (19) for the complete mixing case in Figure 11.

The two cases are very similar. A borehole log responding to concentration cannot distinguish between the two cases because both produce a constant concentration throughout the hole. At low flows, the concentration ratio for

both cases increases linearly with $Q/\lambda V$. At intermediate flows, the dependence is logarithmic, with the mixing model producing concentrations higher than the continuous entry model. At very high flows the entry concentration will be observed.

Discussion

We have developed a few simple models to describe the radon concentration within a hole, using different assumptions for mixing and for entry point locations. The resulting expressions for three borehole models are given in Eqs. (16), (19) and (23), where the borehole concentration can be expressed as a function of the entering concentration C_0 . The concentration ratio is generally dependent only upon the volumetric flow rate Q and the borehole volume V . For the single entry point with no mixing, the flow rate can be inferred from a log that is proportional to the concentration. For the mixing and uniform entry cases, the concentration and the count rate are constant throughout the volume V , so that C_0 can be estimated, or, conversely, C_0 must be independently established if Q is to be estimated. We have also pointed out that the case of complete mixing cannot be distinguished from the case of continuous uniform fluid entry into the hole. Independent hydrological evidence is needed to distinguish them.

The thin crack expression for C_0 of Eq. (10) can be merged with any of the four borehole models to provide an expression for the borehole concentration in terms of the flow and source parameters. As an example, the thin crack result of eq. (10) is combined with the mixing model of Eq. (19) to give

$$\bar{C} = \frac{EQ}{\lambda^2 hV} \left(1 - e^{-\lambda V/Q} \right) \left(1 - e^{-\lambda \pi h(b^2 - a^2)/Q} \right) \quad (24)$$

For various limits of the groundwater inflow rate, Eq. (24) yields quite different expressions for the radon concentration in a borehole. Table 4 summarizes the four conditions where the flow is either very high or very low with respect to the product of the radon decay time and the radon source and borehole volumes. As the table shows, if the fluid spends considerable time in both the source area ($Q < \lambda V_c$) and within the borehole ($Q < \lambda V$), then

$$\bar{C} \approx \frac{EQ}{\lambda^2 hV} \quad (25)$$

and radon concentration in the borehole increases linearly with flow rate. We have already stated that Q/λ will be small compared with the source volume in the crack, making the term in the second pair of brackets in Eq. (24) approximately equal to one. For the larger borehole, Q/λ will also be small compared with the mixing volume (see Table 3 for values of λV), and hence this result seems appropriate for the Stripa results of Fig. 4, since the flow values are reasonable and the concentration increases linearly with the flow rate. It must be emphasized however, that this linear dependence upon Q is really a result of long fluid residence times within the borehole and source area.

Otherwise, if the fluid spends only a fraction of the radon half-life in either of these volumes, the concentration either remains constant or depends inversely upon Q . In the right-hand column of Table 4, where $Q > \lambda V$, the flow is so high that the borehole does not affect the concentration, which maintains its entry point levels.

FLOW RATE ESTIMATES FROM GAMMA-RAY BOREHOLE LOGS

The mixing effects appeared to be limited to the larger boreholes of 127 mm diameter, as judged by the character of the gamma-ray logs acquired in the larger holes. We now discuss results from smaller boreholes of 76 mm diameter, where monotonic decay of count rate with distance from a fluid entry point was observed in a number of cases. For these cases, eqn. 16 and the subsequent discussion applies.

Figure 10 shows results from borehole S2, which is collared in the main experimental drift at 339 m subsurface and dips at a 30-degree angle with the horizontal. The gamma logs in Fig. 10 were obtained over a 1-1/2 year time period. Two sharp peaks are present on all three gamma logs at 25 m and 38 m depth. These peaks are attributed to uranium decay products in rock because of their narrow half-width and because of their additive contribution to the three logs. The broad, time-varying increase over the interval 24-32m is attributed to radon in water.

Hydrological and fracture data from S2 have been reported by Gale and Witherspoon (1979), and injection test results proportional to permeability are shown in the right hand side of Figure 10. The radon increases depicted in the figures occur within the permeable zones, and decay above them. The upper permeable zones do not appear to be contributing radon, however. We believe that radon-charged groundwater is entering the boreholes at the lower permeable zones, moving up the holes, and exiting near the collars.

The logarithmic plot of count rate in Fig. 10 establishes flow-rate estimates of 3.0 and 11.5 liters per day over the indicated intervals for the latter two logs, using eqn. 16. The sustained level above the decay line in the upper eight meters of log may indicate an additional contribution

from the upper permeable zone. The changes of flow rate with time are related to changes in the hydrological regime induced by thermomechanical and hydrological experiments underway in adjacent drifts. In particular, the higher flow recorded on the November 1979 log is related to the packing off of a copiously flowing borehole located 35 m away. No measurements of outflow were made for the first two logs, but in February, 1980 the outflow at the collar of S2 was determined to be 13.4 liters per day, a very encouraging check with the 11.5 liters per day estimate from the gamma-ray decay.

Gamma-ray data and a geological column for surface borehole SBH-1 (refer to Figure 1 for location) are shown in Figure 11. This hole, which has a total length of 380 m, was drilled at a 45° angle from the horizontal, and the depths given in Fig. 11 are slant depths. A second gamma log, not shown here, was identical to the first except that the sharp decrease at 78 m occurred instead at 90 m. In each case the decrease occurred as the probe entered the standing water column in the borehole.

We interpret the log behavior above the water level to be caused by the introduction of radon into the wellbore by groundwater infiltrating into the open hole and trickling down the lower side. The higher count rate observed above the water level is attributed to the fact that radon is more soluble on a volumetric basis in air than in water. Water entering the hole gradually releases its radon to the air as it trickles down the hole. The steady release during trickle flow could be coupled with diffusional loss of radon upwards and out the open collar to cause the count rate gradient observed above the water column. Once the radon-charged water reaches the top of the water column, the count rate drops because the water has already released most of its radon.

The radon-charged water trickling down the open bore does not mask the spikes caused by uraniferous zones, nor does it mask the count decrease observed at the granite-leptite contact near 52.5 m. The count rate in leptite above the water level is constant because the leptite does not contribute radon-charged water. This does not exclude the possibility that water is entering through fractures in the leptite, however, because above the water column the count rate would not be affected.

The individual data points are based on spectral analyses of core samples, adjusted for calibration of the probe. Two of the features are ascribed to geological effects. The step-like increase of about 140 cps at 208 m at the granite-leptite contact checks quite nicely with spectral data which shows a computed count difference of 150 cps between granite and leptite (compare samples 148.65 m with 232.85 m, etc.). A second feature, a reduced count rate at 280 m, is associated with a 2.5 m intercept of "greenstone," which may be a leptite xenolith.

Most important in SBH1, however, is the excess of actual counts above the expected rate indicated by the five data points. In the leptite, the gamma log should be indicating 40 to 50 cps; instead, it registers almost 200 cps. In the granite, 200 cps is expected, based upon the logging in underground holes as well as the computed response; instead, the count rate is in excess of 300 cps. These excess rates are attributed to radon transported downward in the water column and eventually exiting the borehole at depths where fracture zones occur. The most prominent of these fracture zones occurs at about 320 m, below which depth the count rate declines to the expected value of 200 cps. The gradual decline below 325 m indicates some further loss of water below 320 m, as discussed below.

This interpretation of fluid movement is consistent with other data available on SBH-1. The fractures observed in core and the geophysical logs (Nelson et al., 1979) reveal the presence of an 8 m interval of fractured rock centered at 320 m. A shift in the static temperature gradient at that depth also convincingly indicates that outflow occurs. And finally, water pressure measurements (Gale and Witherspoon, 1979, Figure 7) show that in situ pressure is less than hydrostatic below a depth of 150 m.

The downward flow rate must be quite high above 325 m in SBH-1 because no gradient in the gamma log is discernible. As a consequence, only a minimum flow rate can be established using the concept presented in Fig 8. Multiplying the length parameters in that figure by a factor of 20 provides an assessment of the SBH-1 case where we observe no gradient over the 240 m column of flowing water. The minimum flow rate down to 325 m in SBH-1 is estimated to be 1000 liters per day.

Below 325 m the flow rate reduces to about 50 liters per day with most of it existing at another fracture zone at 337 m, as shown in the detailed gamma log of Fig. 12. Hydrological and geophysical data also indicated in Fig. 12 confirm the location of the fracture zone. Below 337 m the flow reduces to 3 liters per day. Measurement sensitivity does not permit detection of the exodus of this small amount of flow.

In sum, the downward flow rate in SBH-1 under static open hole conditions is in excess of 1000 liters per day. Over 90% of this flow exits the hole at a fracture zone at 320 m. Less than 10% of the flow exits at a second fracture at 337 m.

IMPLICATIONS FOR RADON EMANATION AND URANIUM DISTRIBUTION

The thin crack model and the measurements of radon activity in the M3 water allow us to make some crude estimates of the aperture h of a permeable fracture. Although an expression such as eqn. 25 could be used if the flow rate Q and the concentration after mixing were known, a more direct approach follows from eqn. 14,

$$h = E/\lambda C_o \quad (26)$$

where E , the surface emanating power derived from the thin crack model, must be related to the volumetric emanating power E_v which gives the radon produced per volume (or weight) of rock. Emanation values E_v measured on Stripa core samples which were air-dried at room temperature are tabulated in Table 1. There are 21 samples for which the average value of E_v is 1.42 pCi/gm, equivalent to

$$E_v = 1.42 \times 10^{-12} (\text{Ci/gm}) \times 2.63 (\text{gm/cm}^3) \times 3197 \times 10^{12} (\text{dis./day/Ci})$$

$$E_v = 12 \times 10^3 \text{ radon atoms per day per cm}^3 \text{ rock}$$

An estimate of diffusion distance supplied by I. Neretniks (pers. comm. 1980) suggests that a square centimeter of fracture surface has an effective volume extending about 2 cm from either side of the fracture. This estimate of the rock volume contributing radon to a unit area of fracture surface implies that $E/E_v \sim 4 \text{ cm}^3/\text{cm}^2$. Coupled with the laboratory measurements of E_v , we find that $E \sim 48 \times 10^3$ radon atoms per day per cm^2 of fracture surface.

An estimate of the entering concentration C_0 can be based on the measured activity of the M3 water (Table 2) of about 1 μCi per liter, which by

$$N = \frac{1}{\lambda} \frac{dN}{dt}$$

converts to 18×10^6 atoms per cm^3 . A factor 3197×10^{12} was used to convert activity in Ci to number of disintegrations per day.

Use of these values for E and C_0 in eqn. 26 results in a value of 0.15 mm for fracture aperture in borehole M3. This value is consistent with injection test results by Gale (1981), who computed fracture apertures as large as 0.2 mm by attributing all outflow from a 2 m interval to a single fracture. Hence the computation demonstrates that the emanation values and the fracture-flow model for radon transport are reasonable.

Our findings indicate that the high radon concentration in Stripa groundwater is satisfactorily explained by a combination of high uranium content and thin fracture aperture. These results also imply that other igneous rocks with similar fracture characteristics will produce radon concentrations differing from that at Stripa in proportion to the fraction of the uranium present. If so, then many sites must exist where the radon concentration is 10% or more of the Stripa concentrations, although there is little documentation to support this.

The fraction of radon atoms formed which escape from the rock is sometimes called the coefficient of emanation (Tanner, 1978). The data in Table 1 show that 0.10 is quite a reasonable value for the fractional emanation coefficient, which conforms well to the estimates of escape to production ratio obtained by Barretto et al. (1972). With reference to this data base, the Stripa samples do not appear to be unusual in fractional emanation.

Another possibility is that radon emanation is extraordinarily high along permeable fractures and that these surfaces were not represented in the core samples reported in Table 1. Mobilization and deposition of any member of the U-238 series higher than radon could produce such an effect. Two likely candidates are uranium and radium. Wollenberg et al. (1982) used the fission track-radiographic method to locate and determine the abundance of uranium in uncovered thin sections. They find that uranium is localized in the Stripa granite (quartz monzonite) in three distinct mineralogical associations. In brief, they find that uranium is found concentrated in:

- 1) tiny euhedral opaque grains found usually in chlorite, but also in muscovite-chlorite-sericite filled fractures, and even within quartz or feldspar grains.
- 2) anhedral opaque grains associated with both a quartz-epidote-sericite-filled fracture and with fine carbonate-sericite stringers. The absolute abundance of uranium in this second category is greater than in the first.
- 3) dispersed along chlorite-filled fractures without associated discrete grains.

The number of thin sections examined was small, so no statistical inferences were possible. At the present time, the petrographical evidence indicates that uranium is preferentially disposed along flow paths but the evidence is far from conclusive. However, Wollenberg et al. (1980, 1982) present another line of evidence indicating that uranium is located in sites that can be leached by groundwater rather than in inaccessible sites in accessory minerals. Although the outcrops at Stripa are comparatively fresh as a consequence of glaciation, the uranium content in surface outcrop is depleted (mean of 27 ppm) relative to subsurface samples (values in the range 40 to 45 ppm). The data provide good evidence that uranium sites within the rock favor its leaching by acidic near-surface groundwater.

SUMMARY

At Stripa the groundwater contains sufficient amounts of radon to be observed on the total-count gamma-ray log, over and above the gamma contribution from the rock itself. The radon level is high at Stripa because:

- a) the uranium content in the Stripa granite is high, between 35 and 40 ppm.
- b) the uranium seems to be localized along fractures.
- c) dilution is minimal because the porosity is quite low (<1%), or more precisely, because the fracture aperture is small.
- d) flow rates are high enough to bring in enough radon for detection, yet low enough to provide sufficient residence time in the source volume.

Step-like increases (decreases) in radon levels reflect zones of fluid entry (loss). Examples are the fluid loss zone at 320 m slant depth in SBH-1

(Fig. 11) and the fluid entry zone in borehole S2 (Fig. 10). These examples establish that the radon-charged water provides a flow profile of the hole, showing which zones produce or take water under hydrostatic open hole conditions.

If mixing occurs within a borehole, then the average concentration of radon may depend upon the volumetric flow rate as Q^{-1} , Q^0 , or Q^{+1} , depending upon the ratio of Q/λ to the source and mixing volumes (see Table 4). We mainly see evidence for mixing in the largest (127 mm) diameter holes (refer to Fig. 4), where the inflow rate Q is always low enough that the concentration varies linearly with Q . In this case, concentration is controlled by the residence time in the hole that is less than the radon half-life. Fluid entry from a single fracture followed by mixing cannot be distinguished from uniform fluid entry along the borehole length (refer to Fig. 9 and the corresponding discussion).

If mixing does not occur, and if the flow rate falls within a range of values determined by the radon half-life and the observation length within a hole, then the flow rate can be estimated from the exponential decay of radon activity along the hole (refer to Fig. 8). Flow estimates in boreholes S1 and S2 were both about 11 liters per day, within 2 l/d of the flow measured with the bucket-and-stopwatch method. The downward flow in SBH-1 is so high that the activity level remains constant down to the loss zones; the rate must exceed 1000 l/d. No independent check on the SBH-1 estimate is available.

The natural radioelement distribution at a potential site for radioactive waste storage must be carefully determined during the site characterization phase, to determine levels of the natural radioisotopes in rock and

groundwater. Although not many cases have been documented, high radon levels in groundwater may not be all that unusual in granitic rock sites: a recent groundwater study in Maine by Hess et al. (1980) shows that radon levels in groundwater are higher in granitic than in metamorphic rock, usually above the nanocurie per liter level, with some samples as high as 0.2 μCi per liter. Hence careful baseline studies are necessary, and even then one must remain aware that the natural radioactive component of groundwater may change due to changes in the hydrological regime.

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		Crushed samples				Whole core	
		DBH V-1	N1	SBH-1	SBH-2	N1	N1*
Number of samples		15	1	4	4	55	3
U(ppm)	high	47.4	-	41.3	29.96	67.8	163.8
	mean	39.4	40.8	38.9	17.51	38.7	140.8
	low	12.8	-	35.7	7.76	20.9	115.1
Th(ppm)	high	39.6	-	30.6	31.95	40.7	98.8
	mean	32.9	38.8	29.8	28.80	26.9	87.0
	low	26.4	-	28.6	22.5	9.7	77.5
K(%)	high	4.74	-	4.26	4.01	5.16	4.02
	mean	3.88	3.92	4.01	3.60	4.03	3.84
	low	3.10	-	3.87	2.82	3.10	3.67
Rn-222 emanation	high	0.158	-	0.193	-		
	mean	0.096	0.096	0.164	0.108**		
	low	0.044	-	0.117	-		
Rn-222 (pCi/gm)	high	2.24	-	2.38	-		
	mean	1.28	1.30	2.11	0.84**		
	low	0.38	-	1.61	-		

* three samples were much higher than rest of population.

**one sample only.

Table 1. Uranium, thorium and potassium analyses from gamma-ray spectra on crushed and whole core samples of Stripa quartz monzonite. Radon emanation data given for crushed samples.

Table 2 Radon-222 and radium-226 analyses reported by Fritz et al. (1979),
Tables 1 and 14. Samples designated AT were analyzed by AB
Atomenergi, Sweden; UB, by University of Bath, Great Britain.

Sample number	Location	Sampling Date	Radon-222 ($\mu\text{Ci/l}$)	Radium-226 (pCi/l)
16(AT)	M3	9-21 Sept 77	1.9	n.d.
16(UB)	M3	9-21 Sept 77	1.3	34
17(AT)	410-hole 6.3-50m	9-20 Sept 77	0.48	n.d.
29(UB)	410-hole 376.5-471m	Jan-Mar 78	0.56	40

Table 3 Volume capacity of boreholes, in liters per meter length of
hole. The length and diameter of the gamma ray probe are 2.07 m
and 33 mm, displacing approximately 1.8 liters. The third column
gives the product of λ , the radon decay rate, and V, the volume of
a hole 10 m long of specified diameter.

Hole diameter (mm)	Volume per length (l/m)	$\lambda V(1/\text{day})$
38	1.13	2.0
56	2.46	4.4
76	4.53	8.2
127	12.67	22.8

<u>Flow rate Q vs mixing volume V</u>		
Flow rate vs. crack volume V_c	$Q < \lambda V$	$Q > \lambda V$
$Q < \lambda V_c$	$\frac{E}{\lambda h} \frac{Q}{\lambda V}$	$\frac{E}{\lambda h}$
$Q > \lambda V_c$	$\frac{E}{\lambda h} \frac{V_c}{V}$	$\frac{E}{\lambda h} \frac{\lambda V_c}{Q}$

Table 4. Average radon concentration with mixing on borehole of volume V , after inflow from a thin flat crack of aperture h and volume V_c .

XBL 792-7380C

Figure 1. Plan map of the Stripa site showing rock outcrop, inclined surface boreholes and underground experimental drifts. Mine coordinates given in meters.

XBL 809-2805

Figure 2. Gamma-ray spectrum on crushed core sample from 3.10 - 3.49 m interval in borehole N1. Energy (keV) and isotope identified above prominent peaks. Windows for potassium, uranium and thorium analysis specified by horizontal arrows. Sample contains 44.6 ppm U, 27.0 ppm Th and 4.10% K.

XBL 79I-70

Figure 3 Gamma-ray logs along axis of the full-scale drift. Vertical and horizontal spatial scales are identical except at discontinuity between E8 and EI3. Baseline counting rate (counts per sec) is given above each hole.

Borehole gamma ray logs and water inflow rates in time scale heater holes

XBL 7910-13057

Figure 4 Borehole gamma-ray logs and water inflow rates in 127-mm diameter boreholes.

Sequence of gamma ray logs in hole M3
M3 was dewatered at 0:00 hours

XBL 7910-13056

Figure 5 Sequence of gamma-ray logs in hole M3, which was dewatered at 0:00 hours. Final log was run 20:16 hours:minutes later.

XBL 802-6743Y

Figure 6 Gamma ray activity of water from M3, measured with probe inserted into 60-liter vessel. Count time is 100 seconds. The straight line gives the decay of radon-222.

XBL 802-6794

Figure 7 Thin crack model for radon transport. Water flows radially at a rate Q through a thin crack of aperture h into the borehole. The crack contains a circular source area of outer radius b which produces radon atoms at a rate E .

XBL 802-6790

Figure 8 Ratio of the radon concentration in one-dimensional water flow (logarithmic scale), plotted against distance from the source. The radon decay rate is 0.18 per day. Linear flow velocities range from 0.1 to 10 meters/day. Numbers in parentheses give the equivalent volumetric flow in a 76 mm hole.

XBL 803-6861

Figure 9 Radon concentration in borehole water for two infiltration models.

Values of λV are given in Table 3.

Figure 10 Original gamma-ray logs in borehole S2 for three different dates (left). Logarithmic plot of gamma counts with the 200 cps background and anomalies at 25m and 38m subtracted out (center). The slopes of lines 1 and 2 (fit by eye) indicate flows of 3.0 and 11.5 liters per day, respectively. Injection test data (right) taken from Gale and Witherspoon (Fig. 11, 1979).

XBL 808-7296

Figure 11 Complete gamma log of hole SBH-1, logged in February 1978. The five data points are computed from laboratory spectral gamma analyses of core samples.

Figure 12 Detail of gamma log in SBH-1 over the interval 325-350 m, with 200 cps background subtracted. Half-life distances and volumetric flow estimates are posted next to straight-line segments. Sonic waveform anomalies (Nelson et al, 1979) and straddle packer injection tests (Witherspoon et al., 1980) show possible flow zones.

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