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INTERIM REPORT

Radon-222 Emissions in Ventilation Air Exhausted From Underground Uranium Mines

P. O. Jackson R. W. Perkins L. C. Schwendiman N. A. Wogman J. A. Glissmeyer W. I. Enderlin

September 1979

Prepared for the U.S. Nuclear Regulatory Commission under a Related Services Agreement with the U.S. Department of Energy Contract EY-76-C-06-1830

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
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PNL- 3262 Per 3 this Report

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PACIFIC NORTHWEST LABORATORY

operated by

BATTELLE

for the

UNITED STATES DEPARTMENT OF ENERGY

Under Contract EY-76-C-06-1830

Printed in the United States of America Available from National Technical Information Service United States Department of Commerce 5285 Port Royal Road Springfield, Virginia 22151

Price: Printed Copy \$____*; Microfiche \$3.00

*Pages	NTIS Selling Price
001-025	\$4.00
026-050	\$4.50
051-075	\$5.25
076-100	\$6.00
101-125	\$6.50
126-150	\$7.25
151-175	\$8.00
176-200	\$9.00
201-225	\$9.25
226-250	\$9.50
251-275	\$10.75
276-300	\$11.00

NOTE

This is a revision of an earlier document of the same name and number dated March, 1979. This current revision supersedes and replaces the March document, all copies of which should be destroyed.

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Pacific Northwest Laboratory Richland, Washington 99352

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INTERIM REPORT

ABSTRACT

The ²²²Rn concentration in exhaust air is being measured at underground uranium mines in the Grants, New Mexico area. The objective of the work is to determine relationships between U₃O₈ production and the mine characteristics and practices relative to 222Rn emission. Concentrations in the vent air from two mines ranged from 90 nCi/m³ to 3800 nCi/m³ during a month of observations. Diurnal radon emission patterns were seen from each mine which were inversely related to barometric pressure. The average diurnal emission patterns on weekends when no mining occurred were very similar to those on weekdays during active mining, indicating that the mining activities had little short-term effect on those radon emissions. The radon emission rate from each mine vent showed a correlation with the associated ventilated surface area and the cumulative tons of U_3O_8 extracted there and a higher correlation with cumulative tons of ore extracted. Grab samples of vent air collected at 14 additional mines in the Grants area showed radon concentrations from 7 nCi/m^3 to 21,000 nCi/m^3 . The radon emission rates ranged from 0.01 Ci to 10 Ci per vent per day. Ore production rate estimates were available for seven of the mines and based on a composite of emissions and production of these mines, a tentative average radon emission via ventilation air of 15.9 Ci/ton U₃O₈ was obtained. Aboveground sources of radon at one mine were estimated to release approximately 4% of the radon emitted in ventilation air from that mine. Using this estimate, a total release of 16.6 Ci/ton U₃O₈ is predicted. This is equivalent to 3340 Ci radon per RRY (182 metric tons).* After mine closure the waste piles at that mine are estimated to continue releasing radon at a rate of 13 Ci/yr per RRY.

^{*}RRY = Reference Reactor Year is a 1000 MWe reactor operating for one year. In this report the fuel requirement for the reference reactor year is taken to be 182 metric tons (tonnes) of U_3O_8 . Taking into account milling recovery of 90.5%, the requirement of U_3O_8 in the ore as mined is taken to be 201 metric tons.

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INTRODUCTION

In estimating the radiation exposure to the U. S. public from nuclear power, it is important to look at exposure resulting from all phases of the fuel cycle. In this regard, the Nuclear Regulatory Commission has requested that our laboratory conduct a study to determine radon emissions from underground uranium mining operations. To make the data useful in estimating the impact of nuclear power, it is essential to determine the radon emissions associated with mining the quantity of uranium required for operation of a reference reactor for one year (Reference Reactor Year, RRY*).

A review of the literature on this subject indicated that there are essentially no useful measurements of radon emission from underground uranium operations. We have, therefore, initiated a research program which will determine the relationship between U_3O_8 production and radon emission from uranium mine ventilation exhaust vents. The intent of this program is to firmly establish this relationship for operating uranium mines and to determine how mining operations affect this radon release. The program also calls for determination of particulate and gaseous emissions from the uranium mine ventilation exhaust. However, because of the necessity for obtaining information on radon release rates during a relatively short time frame, we have emphasized the radon measurements and this report deals only with these measurements.

Studies that have been conducted in 1978 were all made in the Grants, New Mexico area where approximately 50% of the underground uranium mining in this country takes place. This is an interim report giving the results of measurements made to date. Although these results represent a significant fraction of the U. S. production, industry averages derived from them are tentative in nature until this survey is completed. This report is a revision of an earlier report carrying the same title and number. This revision is made to conform the report to the currently used value for RRY, to change the format of presentation, and to make some small corrections for consistency.

EXPERIMENTAL

SITE SELECTION

One aspect of this program consisted of studying a relatively few mines in enough detail that it would be possible to predict the radon emissions from the majority of the industry by knowing the characteristics of each mine.

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^{*}RRY = Reference Reactor Year is a 1000 MWe reactor operating for one year. In this report, the fuel requirement for the reference reactor year is taken to be 182 metric tons (tonnes) of U_3O_8 . Taking into account milling recovery is 90.5%, the requirement of U_3O_8 in the ore as mined is taken to be 201 metric tons. (Ref. 1)

To minimize the complexity of data interpretation, it was necessary to select a mine for study that had certain characteristics. Those included:

- 1. A straightforward ventilation system to facilitate interpretation;
- A relatively small number of vents to minimize the field effort and equipment requirements;
- Differing operations at individual vents so that several parameters could be examined;
- Ventilation air sufficiently dry that dust sampling would be practical;
- 5. A mine of a major producer with typical operating procedures; and
- 6. A relatively high production rate.

Mine 1

A schematic map of the first mine selected (Mine 1) is shown in Figure 1. This mine has five upcast vents. All of the incoming air comes down the shaft. The working areas are not contiguous. Air passes via a lower level haulage way directly into each area without significant contamination from the others. It then flows up through each working area and from there into the exhaust vent located at the site. This is a relatively young mine which was opened in 1970. The mine used a two-shift operation with only maintenance work on Saturdays. Blasting occurs at the lunch times, between the day and swing shifts, and at midnight. Some of the characteristics of this mine were obtained from the superintendent, geologists, and ventilation technicians and are listed below.

Vent 1

Vent 1 uses a small fan to maintain a net inflow of air into an abandoned worked-out area. The ore in this portion of the mine had 4 to 5% moisture by weight. The rock was described a; "fairly tight" with fine fracture channels. Some caving and roof fall have taken place in this area. It is not sealed with bulkheads. The ventilation rate was approximately 9700 cfm and the vent air contained essentially no entrained water.

Vent 2

The exhausted area is the wettest of the mine. The moisture content was about 8%. Some water flows from disturbed surfaces, but the total was only about 35 gal/min for the entire area. The ore body is highly frac-

tured and has a low permeability and a high porosity on the average. The ventilation air contains a small amount of entrained water. The vent flow rate is about 30,000 cfm. Stope mining activities are characteristic of a fully developed mine. The area of the mine is relatively small.

Vent 3

The Vent 3 area is the largest of the mine. Mining activities are essentially the same as at Vent 2. It is drier than the Vent 2 area, higher, and less faulted. It contains the most extensive array of working surfaces in the mine. The ventilation rate is approximately 80,000 cfm with no entrained water.

Vent 4

The Vent 4 area is a new mining area in early stages of development. No stope mining was started until the summer of 1978. Most of the surface area consists of drifts. The ore body is thick and complex in overlapping layers which were faulted and fractured after geological deposition. It is fairly dry with a moisture content less than 5%. A single large stope is being formed currently. Drift development is also progressing. The vent flow is about 33,000 cfm with no entrained water.

Vent 5

Vent 5 exhausts a thin sandy deposit. It is a new area in a more advanced state of development than that of Vent 4. The vent air contains much entrained water which is thrown in a coarse spray. The water is not from mining areas but enters the vent from groundwater near the surface. The vent flow is about 38,000 cfm.

Mine 2

A second mine (Mine 2) was selected because of its different characteristics. The second mine is an older mine than Mine 1. It is much deeper and wetter. A large part of the mine consists of abandoned workings which are bulkheaded to prevent back flow into other working areas which are contiguous. Certain areas of the mine have been backfilled. A diagram of this mine was not attempted because of its complexity. The second mine has five vents. Vents 1 through 4 are upcast and Vent 5 is downcast. The air path is from the shaft to Vents 1, 2, 3 and 4, and from Vent 5 to the Vent 4

area. Work is performed as a three-shift operation. However, graveyard shift is used for maintenance, duct installation, and hauling muck. A limited amount of drilling and blasting occurs. About 10% of the work force is on the graveyard shift. The average ore grade is more than double that at Mine I but the production rate of ore is less. The normal blasting schedule is at lunch breaks and at shift changes. The geology of individual ventilated areas was not available, but a general description of the areas is as follows:

Vent 1

Vent 1 exhausts an old abandoned area. The area is bulkheaded to reduce backflow of air. The vent flow was approximately 18,000 cfm and the air was free of water droplets. The area is contiguous with the old, worked-out areas of Vent 2 and cannot be distinguished from it.

Vent 2

Vent 2 exhausts an extremely large area of old workings which are extensively bulkheaded. Parts of the area have been backfilled with sand. There are adjacent areas exhausted by Vent 2 in which active mining is progressing to remove residual ore from some of the old workings. The ventilation rate was approximately 76,000 cfm. Ventilation air contained a small amount of entrained water with a high content of a fine black silt.

Vent 3

Air from Vent 3 passes through a small newly developed area with active mining. The ventilation rate was approximately 29,000 cfm and very little entrained water was present.

Vent 4

Vent 4 exhausts a relatively large complex area being actively mined. It is in a mature state of development. Air flow was into the area from the shaft as well as from a downcast vent. The vent flow was about 100,000 cfm. The exhaust air contained an extremely heavy spray of entrained water droplets.

The air path at Mine 2 was not sufficiently isolated to preclude a partial cross-contamination between areas. Data interpretation was complicated because of relatively frequent vent fan shutdowns.

Air from all vents was very humid and had an essentially constant temperature of between 75°F and 80°F. The activities at the various ventila-

tion areas of Mine I are representative of the stages of mine development. Thus, the areas exhausted by the five vents can be considered as a set of mines in various developmental stages operated with the same general mining practices.

In addition to the extensive continuous monitoring of Mines 1 and 2, a program of grab sampling at many other mine vents was conducted.

FIRST FIELD TRIP

An initial field trip to Mine I was made to evaluate procedures and equipment and also to obtain a preliminary set of measurements. A one-week sampling trip to the Grants, New Mexico area was made in late March, 1978. A mobile laboratory van was stationed at Mine 1, Vent 3. One of the first tasks of this program was the adaptation and development of equipment suitable for monitoring radon concentrations. Several approaches were considered and three were evaluated on this field trip. Internal scintillation flasks, charcoal traps for collecting radon, and a prototype flow-through radon counter were compared. After evaluating the performance of these instruments, it was decided that the internal scintillation flasks were the most advantageous for simultaneously monitoring a number of mine vents. They are easily transported in the field, relatively rugged, could be constructed inexpensively in relatively large numbers, did not require cooling, and could be monitored in the field. Their greatest disadvantage was that they were essentially grab sampling instruments. However, by using a sequential air sampler to continuously collect samples in the field, each day's emission could be divided into increments useful for evaluation of release patterns. Five sequential bag samplers were purchased following the first trip. These samplers were modified to permit vent sampling. Two flowthrough scintillation flask constant radon monitors were subsequently purchased but not in time for most of the measurements.

In addition to radon measurements, high volume dust samples and cascade impactor stages were taken on the first trip. A small array of soil samples was also collected at each vent. The short-lived radon daughters, as well as ²³⁸U and ²¹⁰Po, were measured in particulates. Aluminum, vanadium, and uranium were also measured using neutron activation techniques. The results of these measurements will be covered in a separate report.

SECOND FIELD TRIP

A second field trip to the Grants, New Mexico area was made from August 1, 1978 to November 1, 1978. Sequential samplers were installed at the five vents of Mine 1. A field laboratory was stationed between Mines 1 and 2. Radon concentrations were monitored continuously at Mine 1 until the second week of September. At that time, the sequential samplers were moved and installed at Mine 2. Measurements were taken there until the second week in October.

Between the middle of September and the end of October, grab samples were collected at 14 other mines in the Grants area.

A short stopover at the Denver Federal Center to use the ²²²Rn calibration facilities of the Bureau of Mines was made on the second field trip. SAMPLING

Sequential Sampler

To collect samples of vent air in a continuous sequence, five modified sequential bag samplers were employed.* These samplers were programmed to pump approximately 2 liters of air into a tedlar plastic bag in four hours before switching to the next bag. Six bags were used per 24 hours' sampling. Once per day, the bags were clamped shut and taken to a transfer point. Transfer to evacuated scintillation flasks was made in the field at those vents having available power for the pump. In other cases, filled bags were replaced with evacuated bags and transported to a field laboratory for transfer of the vent air sample.

The sequential sampler modifications are detailed in Figure 2. Inlet air flowed through a 3" flex tube plastic duct connected by a mounting fixture to an 8" x 8" HEPA filter attached to the plenum cover of the sequential sampler. Filtered air passed into the sample pump plenum through a hole cut in the cover. The air path was across the plenum to its periphery where it exited from beneath a plastic film skirt. The skirt was fitted around the drum and plenum. It served to prevent drafts from diluting sampled air.

Air from the mine vents had an exit velocity of 1000 to 5000 ft/min. A flexible duct was attached at the top of a vent with its open end facing the air stream. The duct carried air through a water separator and then

^{*}AQS-II (25 % model), Environmental Measurements, Inc., Mountain View, California.

into the HEPA filter. The water separators, constructed from 5-gallon metal cans, were used as shown in Figure 3. For small quantities of entrained water, the cap was removed daily for draining. When the entrained water would fill the can in less than a day, the cap was removed and the can inserted into a 6" deep pan of water. Excess water constantly overflowed the pan while the 6" deep water seal prevented escape of the air and radon. A diagram of the entire air collection system is shown in Figure 4.

The distribution coefficient for radon into water from an equal volume of air at ambient weather temperatures is about 0.25 (Ref. 2). Since the volume fraction of water in the flowing stream is much smaller than 0.01, loss of radon through the water seal was considered negligible.

A mechanical vacuum pump and manifold system was used to transfer air from the tedlar bags to the scintillation flasks. After the flask and manifold lines were evacuated, the pump was valved off and the bag opened to the system. After an interval of 15-20 seconds, the flask was valved shut. The bag was removed and attached to a battery-powered pump which evacuated it prior to re-use.

On completion of the alpha particle counting, the scintillation flasks were attached to a manifold for evacuation with the mechanical pump. When the flasks contained less than 5 torr residual pressure, they were refilled with room air which had been passed through activated charcoal. They were then re-evacuated and refilled. After a final evacuation, the flasks were stored overnight. The residual pressure in each flask was then measured. Flasks which leaked more than 60 torr overnight were not reused until the vacuum leak was repaired. Normally, flasks leaked less than 20 torr overnight.

Grab Samples

Grab samples were collected directly into evacuated scintillation flasks. Two flasks in sequence were filled from the exhaust stream at the vent exit. A fiberglass filter was attached to the inlet to remove particulates and entrained moisture droplets. A second set of duplicates was taken at each vent on another day.

RADON ANALYSIS

Two techniques were used to determine the concentration of radon in mine vent air on the second field trip. Internally coated scintillation flasks were used for most of the determinations. Commercially available scintillation flasks* were used for cross-calibration purposes. These flasks have an internal volume of 95 ml and a nominal efficiency of 5 c/m per pCi of radon. For field work, forty large volume flasks were constructed of 6" I.D. plexiglass tubing. The internal volume was 1136 ml. The scintillator, a mixture of zinc sulfide television** phosphor and clear coil dope was spraypainted to internal surfaces. Exterior surfaces were coated with glossy white aerosol spray paint. These flasks were cross-calibrated with the commercial units at the facilities of the Bureau of Mines in Denver, Colorado, and again in the field. The commercial units were cross-calibrated with identical units which had been individually calibrated with radon from a ²²⁶Ra standard by staff members of the Bureau of Mines. A discussion of these calibration measurements, as well as other calibrations and quality assurance data, will be found in Appendix A.

After evacuation and filling, scintillation flasks were stored for five hours or longer to permit radioactive equilibrium between radon and its daughters. The scintillations were detected with photomultiplier tubes in light-tight boxes. At first, the flasks were coupled to the photomultiplier tube with optical coupling compound. Later it was decided to remove the compound to reduce strain on the detector components during decoupling. The system was recalibrated after removing the compound. Eight photomultiplier tubes were used for detection. Each was calibrated to correct for small sensitivity differences between them. Pulses from the photomultipliers were detected with commercial preamp-amplifier-discrimination modules and stored on scalers. Up to eight detectors could be monitored simultaneously with the unit illustrated in Figure 5. A 2" photomultiplier tube is used with commercial flasks and 5" tubes are used for the shop-made flasks.

A second measuring technique was used to evaluate the rapidity of changes in radon concentrations at Mine 1, Vent 4. This unit was a commercial continuous flowing radon monitor.*** The monitor consists of a flow-through

^{*} LAC-2 radon counter. Johnston Laboratories, Inc., 3 Industry Lane, Cockeysville, Maryland

^{**} GE RETMA P-11 Type 118-2-4, General Electric Co., Cleveland, Ohio ***Radon Gas Monitor RGM-1, Eberline Instrument Corp., Santa Fe, New Mexico

scintillation flask with a preset timer-scaler and printer in a portable case. The scaler was preset to an interval of 20 minutes after which the stored count and clock time were printed. The count then cleared automatically and restarted. This unit was placed in a weatherproof plywood box. A sample line of copper tubing connected with flexible PVC plastic tubing was used to bring air from the plenum of a sequential sampling unit. This procedure assured a sample free of entrained water. The inlet filter paper was changed daily when the cycle was restarted. The instrument was used for a limited investigation of short-term variations in radon concentrations.

VENT FLOW RATES

Vent flow rates were measured using a pitot tube* with an oil-filled manometer serving as a differential pressure gauge. Where possible, the flow was measured once using an 8-point traverse along a single diameter. When only a single pitot tube hole was available in a vent, a 4-point traverse from the vent wall to its axis was used. Since the manometer had no damping capability, readings at each traverse point were observed for 1-2 minutes to obtain an estimate of the average differential pressure. Because of accidental damage to the equipment, at some vents flow rate measurements have not been made at this time. The mine operator's flow measurements were used when available in those vents we could not measure.

CALCULATION/DATA PROCESSING

The concentration of radon in vent air was calculated using the formula: $nCi/m^3 =$

(net c/m)(counting efficiency)⁻¹(photomultiplier factor)⁻¹ $\left(e^{\lambda}Rn^{(T}ct^{-T}s)\right)$

where counting efficiency is c/m per nCi radon, $T_{\rm ct}$ and $T_{\rm s}$ are the midtimes of the counting and sampling intervals. The radon emission rate for each vent was then determined using the measured vent flow:

$$nCi/m^3 \times m^3/hr \div 10^9 = Ci/hr$$
.

Each calculated emission rate for the constantly monitored vents was placed in an array according to the sampled interval. An array consisted

^{*}Dwyer Instruments Model 160-24P, Dwyer Instrument Co., Michigan City, Indiana

of 24 hourly elements for each day of a calendar month for each vent. Samples were collected for 4-hour intervals, six being taken per day. The intervals were not identical since they depended on the restart time of the sampler each day. Since 24 hourly data points were desired, each 4-hour sample result was assigned to the four array elements corresponding to the sample interval (with minutes truncated). If a vent was sampled during two months, the data were divided into two arrays.

The monthly mean emission rate and the corresponding standard deviation for each hour was determined for each vent. Array elements were also sorted into two groups of weekdays and weekends and hourly averages and standard deviations were calculated. A weekday was defined as Monday 7:00 a.m. to Saturday 7:00 a.m.; weekends were the remaining time. Because of malfunctions of the sampling equipment, the data arrays are incomplete. In addition, measurements taken during known vent shutdowns were not included.

DISCUSSION OF RESULTS

The various measurements that have been made during the past year provide a basis for estimating radon emission as a function of production rate of $\rm U_3O_8$. They also provide an initial basis for determining relationships between radon emission and the nature of the uranium mine. In some initial observations made at uranium Mine 1 during March, 1978, we determined that there was very little relationship between the rate of radon emission from a mine vent and the activities within the mine. These observations were based on the analyses of several grab samples of ventilation air from mine Vent 3 taken over a three-day period and relating the radon concentrations to working schedules and blasting activities in the mine.

As shown in Figure 6, the radon concentrations did not seem to be significantly affected by blasting activities or by whether or not active mining was taking place. The only significant short-term variation in radon concentrations in the exhaust air occurred during a period when one of the ventilation fans for the vent was inadvertently turned off for a 24-hour period. During this period of lower flow rate, the radon concentrations were higher. Since emission rates are the product of concentration and vent flow, these changes would compensate, yielding a relatively constant radon emission rate.

The vent flow was not measured during this interval but was estimated by the mining ventilation engineer to be about 3/4 of the flow with both fans operating.

During the fall of 1978, the radon concentrations in ventilation exhaust air from the five vents of Mine I were measured over consecutive 4-hour intervals for approximately one month. Based on a study of flow rates at one vent, all flow rates were assumed to be constant (see Appendix A).

After correction for the measured vent flow, each calculated emission rate for the constantly monitored vents was placed in an array according to the sample interval. An array consisted of 24 hourly elements for each day of the calendar month for each vent. Each result was assigned to the four array elements corresponding to the sampling period (with minutes truncated). The results for each hour were averaged over the month. The intervals were not identical since they depended on the restart time of the sampler each day.

Because of the variation in sampling intervals, the averages tend to reflect more details of emission patterns than would the use of fixed 4-hour intervals. These results are summarized in Figure 7. These emission rates show a distinct diurnal variation and an inverse correlation with the similarly averaged barometric pressure measurements which are also plotted in Figure 7.

To further determine if there were any correlation between mining activities and radon emission rates, the latter were plotted for weekday periods (defined as 7 a.m. Monday to 7 a.m. Saturday), along with the weekend periods in Figure 8. Emission patterns for these two periods are essentially the same and thus indicate that the variations during each day are predominantly not from mining activities. A similar diurnal variation of the ²²²Rn concentrations measured underground at an inactive uranium mine has been reported by Franklin (Ref. 3).

After making a second series of observations at Mine 2, a similar comparison of the diurnal variation in radon emission rates on weekdays versus weekends was made and is shown in Figure 9. While the basic diurnal patterns for weekdays and weekends remain similar, there are significant variations. It is believed that the differences are due to the fact that relatively few samples were taken on weekends at Mine 2 and therefore incidental variations in barometric pressures and measurement errors may have had a more pronounced

effect. The variation of the measured concentrations encountered at these mines is shown in Table I. It can be seen that the maximum variation was $\pm 30\%$ (of which measurement errors represented about 8%).

As shown in Figure 7, we have established that there is a good relationship between radon emission and barometric pressure which shows a diurnal variation. Since the diurnal variation shown in this figure is actually based on 4-hour integrated samples, we have made measurements to more carefully define the relationship between barometric pressure and radon emission. This was accomplished by use of a continuous radon monitor* which recorded radon emission over 20-minute intervals. The relative concentrations for Vent 4 at Mine 1 are shown in Figure 10 along with the observed barometric pressure variations. It is clear that there is a much more definite relationship between the daily changes in barometric pressure and the radon emission than could be demonstrated with the longer integration times.

Franklin (Ref. 4) has reported several significant local variations in radon concentrations at points underground from blasting and slushing operations. These local concentration changes were apparently masked in our studies when integrated with the large steady-state source from normal exhalation from the mine surfaces.

The radon emission rates from Mines 1 and 2, which include individual averaged emissions from each of the vents, are summarized in Table II. In these two mines, the radon concentrations in the ventilation exhaust air varied by about a factor of 10 while the total radon emitted per day from these vents varied by about 30-fold.

In Table II we have calculated the curies of radon per ton of U_3O_8 mined from the areas exhausted by each vent of Mines 1 and 2 based on typical production data for each ventilated area which was provided by the mine operators. Since these areas are in various stages of development, including abandoned areas with no active mining, the relationship between daily radon emission rates and U_3O_8 production rates does not show a high degree of correlation. However, the total curies per ton summed over the entire mine in each case is a best estimate of the radon emissions for that mine in its current state of operation. We have shown in Table II that the radon emissions are 5.1 and 14.0 Ci/ton of U_3O_8 for Mines 1 and 2, respectively. There is approximately a factor of 2.5 difference in this value for the two uranium mines. This is

^{*}Radon Gas Monitor RGM-1, Eberline Instrument Corp., Santa Fe, NM

a fairly wide difference and does indicate that the older mines will have higher emissions. It will be shown later in our modeling considerations that a relationship does appear to exist between the cumulative tons of ore which have been extracted from a given mine and current radon emission rates.

In Table III we have summarized the relationship between radon emission and U_3O_8 extracted in terms of curies per ton of ore and curies per ton of U_3O_8 for seven uranium mines in the Grants, New Mexico area. These radon measurements which were made by mine operators in 1976 also show a very wide range in radon emission per ton of U_3O_8 extracted from the various mines.

In our current study, we also measured radon emission rates from these mines with the exception of a single vent at Mine 5 which we were unable to sample. Since there are thirteen vents in total at this mine, an estimate of the emission rate of the omitted vent was made. The average radon concentration of the other vents and the flow rate for the missing vent were used (see Table IV). Mines 1 and 2 are those we have already discussed, and the measurements represent the averages of large numbers of samples and more recent production data. For the remaining vents, the production data from Table III have been used. The emission rates for the other five mines are based on averages of the grab sample results at each vent. Because of the limited variation of observed concentrations at Mines 1 and 2, these averages are believed to provide a good indication of emission rates. The current values are generally higher than those observed previously (Table III) and may reflect to some extent the greater maturity of the mines involved. The composite of the seven mines' radon emission in terms of curies per ton of U_3O_8 from these measurements is 15.9. A wide variation in radon release per unit of U₃O₈ production was again observed. This very wide variation makes it rather difficult to develop a generic relationship between the rate of U_3O_8 extraction and radon released to the atmosphere. We will, however, show how this relationship can probably be improved substantially by relating the cumulative production from the mine to the current radon emission rates.

STUDIES CURRENTLY IN PROGRESS

The maximum variation of radon emission rates observed at Mines 1 and 2 was about a factor of 2. Most of the 4-hour integrated samples had a relative standard deviation of less than 20% during about a month of observation.

Since this variation was highly correlated with the barometric pressure, we estimate that we should be able to take a few grab samples from each exhaust vent of active uranium mines and use corrections from the associated barometric pressures to estimate the average of daily emission rate to within about 5% to 10%. We are planning to continue the continuous measurement of radon emissions from vents of several mines in order to derive the functional relationships between emission rates and barometric pressure and to establish the variability of such predictions.

The sampling effort is now well underway and we have collected grab samples from 70 vents at 14 uranium mines in the Grants, New Mexico area. These should represent ventilation exhaust from more than 30% of the underground uranium mines in the United States. The concentrations of radon from these vents vary from 7 to 22,000 pCi/ ℓ and are recorded along with flow rate measurements which are available to date in Appendix B.

As these data are accumulated, we will be able to obtain a much better estimate of the radon emitted per ton of $\rm U_3O_8$ in underground mining operations. We expect to extend our radon measurements at mine vents to include approximately 90% of the U. S. uranium production from underground mining during the next year. With this information and data available on both current and cumulative production of uranium ore and $\rm U_3O_8$, we will have a basis to derive with good precision the current Ci/RRY from the industry, and we expect to be able to develop generic models which provide a good indication of the radon emission which can be expected as mines develop through various stages of maturity in the production of uranium ore.

MODELING OF RADON EMISSION RATES

As indicated previously, the relationship between radon emission and production of U_3O_8 does not show a high degree of correlation (see Table II). We have, therefore, considered the configuration of the mines, production practices, and other factors which may contribute to radon emission rates. In Table V we have shown radon emission rates relative to the total surface area ventilated by various exhaust fans in Mine 1, radon emission relative to the cumulative tons of ore which had been mined from Mines 1 and 2, and relative to the cumulative tons of U_3D_8 which had been extracted from these

mines. All of this information was graciously provided by the mine operators and provided a basis for determining what relationships exist.

When considering the data in Table V, a description of the essential elements controlling radon emissions may prove helpful. The radon emission from a rock surface can be considered as proportional to the product of the surface area, the ²²⁶Ra content of the rock, and the fraction of radon atoms emanated from the surface before decaying. A correction for the fraction of radon which decays underground because of trapping in poorly ventilated or bulkheaded areas must be made when predicting vent emissions. All parameters such as moisture content, mining practices, porosity or permeability may be regarded as modifiers of the basic factors.

It is possible to consider the volume of a mine as roughly proportional to its surface for geometries having a fixed surface-to-volume ratio. The volume, in turn, is proportional to the cumulative tons of rock extracted from the mine.

As indicated in Table V and as shown in Figure 11, there seems to be a reasonably good correlation between radon emission and the surface area of the mine. A linear regression fit was made to the data points in Figure 11 and the square of the correlation coefficients was 0.98. The data in Table V, however, do show a range of 2.25 in the ratio of radon emission to surface area.

The relationship between the radon emission and the cumulative tons of ore which have been extracted from Mine 1 seems to be even closer. In Mine 2, however, this relationship shows a very wide range. To help explain this four-fold variation in radon emission per cumulative ton of ore extracted, it is important to consider something about the mining and ventilation practices. Some of the factors which influence radon emissions are summarized in Table VI.

The areas of Mine 2 ventilated by Vents 1 and 2 had been backfilled to a large extent and are bulkheaded and therefore their radon release could be expected to be attenuated. The relatively high emissions from Vent 4 may be caused by a partial transfer of air from the bulkheaded areas into the Vent 4 air stream. Certainly other parameters may be influencing these emission rates. The important thing to observe is that the total radon emissions

per cumulative ton of ore extracted from Mines 1 and 2 were very similar in spite of significant differences in many of the parameters. Thus, we may expect that as data on cumulative production from other mines becomes available, we may be able to develop a generic model which correctly relates the radon emission to the cumulative tons of uranium ore which have been extracted, with corrections for other parameters which may prove to be significant.

The other relationship shown in Table V is the radon emission per cumulative tons of U_3O_8 . Here the relationship is not as good as that between radon emission and total tons of ore extracted. We suspect that this may be true because the residual ore remaining in the surfaces of the mined-out areas may be almost independent of the initial grade of the ore and, therefore, as mining progresses, one would expect the dominant radon emission to be more nearly proportional to the total quantity of ore removed rather than the uranium content of the material which had been removed.

We are at present accumulating more detailed information about mining practices from a larger number of mines. We expect to correlate variations of the emission rate per cumulative ton of ore from these data as they become available. We expect to investigate these parameters in detail as work continues and thereby develop as realistic a relationship as possible between radon emissions and the related characteristics of the mine and its operations.

PREDICTION OF FUTURE RADON EMISSION RATES

The apparent relationship between the curies of radon emitted from a mine and the cumulative tons of ore extracted offer an approach for predicting future release rates. Once having established the current Ci/RRY the predicted change in that value at any future date would be proportional to the difference between the present cumulative tons of ore extracted from active mines and the cumulative tons at that date. It is important to consider that when a uranium mine closes, the forced ventilation of underground areas ends and the vents may be capped to prevent accidental entry. With proper capping, the radon emission rates from inactive mines are expected to be only a negligible fraction of the rate during active mining. Thus, when a mine ceases operation, the residual radon emission may no longer bear a relationship to the cumulative tons of ore which have been mined. At the

same time, the opening of a new mine of the same daily production rate should add a relatively small radon emission. Thus, in making predictions based on cumulative production statistics, it will be necessary to survey each mine individually, tabulating its cumulative production and its expected lifetime. It will also be necessary to predict the times of opening of new mines and their capacities. This kind of information is not currently being recorded in a central repository. We are at present attempting to locate production statistics sorted into active and inactive mine categories. With such data, a contemporary estimate of radon emission rates may be possible at future dates. Future practices which may or may not include total sealing of old mines will have to be considered in making estimates of radon emission attributed to uranium mining.

ABOVEGROUND RADON SOURCES

A secondary source of radon from underground uranium mining consists of the ore and waste rock which is normally stored aboveground. While the ore is regularly transported to a uranium mill, the waste material is typically used as landfill in an area adjacent to the mine shaft. No measurements of radon from this source were made in this investigation. Approximate radon emission rates at Mine I were estimated from the characteristics of the ore and waste piles. These characteristics were obtained from estimates given by the mine superintendent and by direct observation. Although the stockpile configuration would have a major effect on radon emission rates, the value calculated for those emissions at Mine I is expected to provide an adequate order of magnitude estimate for other mines. Additionally, measurements of stockpiles and waste piles are planned.

Ore Stockpiles: Storage and Handling

At Mine 1, approximately 14,000 tons per month of 0.17% ore is mined. The waste material consists of rock with uranium content below 0.05%. The waste has been deposited in a flat layer approximately 1100' long by 700' wide by 4' thick. On top of the waste material, the ore is piled in rows which are assumed to be triangular in cross-section with dimensions of about 4' high by 8' wide by 500' long. Ore pick-up schedules from the pile depends on factors such as the mill work commitments, mill and mine vacation sched-

ules, and other mine production rates. In general, ore is transported to the mill approximately every two weeks. Thus, just prior to pick-up, about 7,000 tons of ore are on the surface. Using a density of 1.5 tons/yd 3 ,* one computes 126,000 ft 3 of ore on the surface at the time of pick-up. The average surface storage was 3,500 tons. Assuming 226 Ra was in equilibrium with the uranium content, the radon formation rate is:

Radon atoms/yr = $(3500 \text{ tons})(2000 \text{ lb/ton})(454 \text{ gm/lb})(0.0017 \text{ gm } \text{U}_3\text{O}_8/\text{gm} \text{ ore}) \times (0.848 \text{ U/U}_3\text{O}_8)(7.47 \times 10^5 \text{ d/min/gm U})(5.256 \times 10^5 \text{ min/yr})$ (1 atom radon/d ^{238}U) = 1.80 x 1018 Ci radon/yr = $^{12}\text{N}_{Rn} \lambda_{Rn} (2.22 \times 10^{12})^{-1} = (1.80 \times 10^{18} \text{ atoms radon/yr}) \times (1.26 \times 10^{-4} \text{ min}^{-1})(2.22 \times 10^{12} \text{ d/min/Ci})^{-1}$ = 102

Since the maximum thickness of the ore pile is approximately 4 ft and almost 50% of the ore is within 1 ft of the surface of the pile, a diffusion model which assumes an infinite thickness source term is unrealistic. For this estimate we have assumed that 100% of the radon available for diffussion will be emanated, which will give an upper limit to the radon emanation. The fraction available for diffusion from mill waste has been reported as 0.2 (Ref. 5). This yields an estimate of radon emission:

Released Ci radon/yr = $0.2 \times 102 = 20.4$

when divided by the annual U_3O_8 production of 285 tons yields a value of Ci radon/ton $U_3O_8 = \frac{20.4 \text{ Ci radon/yr}}{285 \text{ tons } U_3O_8/\text{yr}} = 0.07$

Since we have assumed that all available radon is emanated as produced from radium decay in the ore pile, it follows that for accounting purposes any further handling would not release additional radon.

^{*}Oensity of ore can vary. We have chosen to use 1.5 tons/yd³ as a nominal, realistic density for our estimates.

Waste Stockpiles

The waste material is stored in a geometry which is amenable to calculation using diffusion models. In this approach, which was used by Nielson et al (Ref. 6) to evaluate emissions from open pit mines, the surface area of the pile was determined, then multiplied by the exhalation rate per unit area, assuming an infinite thickness.

The current quantity of waste from this mine is approximately 1500 tons/month. The density of the ore is approximately 1.5 tons/yd³ and the mine is seven years old. Assuming a life expectancy of 20 years, this waste pile will increase in size by:

 $(1500 \times 12 \text{ tons/yr})(20 - 7 \text{ yr})(18 \text{ ft}^3/\text{ton}) = 4.2 \times 10^6 \text{ ft}^3 \text{ of rock}.$

Assuming a constant depth of 4', as at present, this will represent about 1.0×10^6 ft² of additional waste. Thus, the total area of emanating surface will be 1.77×10^6 ft² after 20 years. The lifetime average would be 8.8×10^5 ft². For this calculation, the effect of the ore stored on top of the waste has been neglected.

The specific exhalation rate used by Nielson was $0.092 \text{ Ci/m}^2/\text{yr}/\% \text{ } U_3 O_8$. Assuming the $U_3 O_8$ content of the waste is one-half the cut-off grade, or 0.025%, the average radon emission rate will be:

Ci
222
Rn/yr = $(8.8 \times 10^5 \text{ ft}^2)(0.0929 \text{ m}^2/\text{ft}^2)(0.092 \text{ Ci/m}^2/\text{yr}/\% \text{ U}_3\text{O}_8)$
 $(0.025\% \text{ U}_3\text{O}_8)$
= 188 Ci/yr , or $0.66 \text{ Ci/ton U}_3\text{O}_8$

Thus, the principal source of aboveground radon will be the waste pile. The total estimated radon from ore, waste, and ore handling would be 0.7 Ci/ton U_3O_8 versus an estimated 5.1 Ci/ton from underground sources. However, Mine 1 has a relatively low emission rate for radon at the present time because of its short mine life. Using the composite of 15.9 Ci/ton for Mines 1-7, the aboveground sources of radon are estimated to produce about 4% of the radon in ventilation air during the operational life of the mine.

Correcting the best estimate of Ci radon emitted from vents for the above-ground sources gives:

$$15.9 + 0.7 = 16.6$$

After Mine I ceases operations, the ore piles will no longer be present and the waste piles will remain indefinitely as a source term unless the waste is removed for processing, or covered to reduce radon release. At that time, for Mine I the total waste pile will produce twice the average lifetime annual radon emission or 2 x 188 Ci/yr = 376 Ci/yr. At the end of a 20-year estimated lifetime, the mine will have produced a total of $(0.78 \text{ tons } U_3O_8/\text{day})(365.25 \text{ days/yr})(20 \text{ yrs}) = 5700 \text{ tons } U_3O_8$. Thus the waste pile will continue to emanate radon at a rate of:

$$\frac{376}{5700}$$
 = 0.066 Ci/yr/ton U₃O₈

BACKGROUND RADON EXHALATION FROM SURFACES COVERED BY WASTES AT THE MINE

The area covered by the waste pile would have a natural background radon emission corresponding to an average uranium content of 0.0004% (Ref. 7). This background radon emission should be subtracted to give the net increase in radon emissions due to mining.

(Area covered by waste $8.8 \times 10^5 \text{ ft}^2$)(0.0929 m²/ft²)(0.092 Ci/m²/yr/% U_3O_8)(0.0004% U_3O_8) = 3.0 Ci/yr, or $\frac{3}{285}$ = 0.01 Ci/ton U_3O_8

After shutdown of the mine, the corresponding value for the area covered by the 20-year waste pile would be 6.0 Ci/yr.

SUMMARY OF RADON RELEASE DURING ACTIVE MINE LIFE

The total and net releases of radon from an underground mine represented in size by production of 285 tons U_3O_8/yr are summarized as follows:

Source	Emission/Ci/ton U ₃ O ₈	Ci/Year		
Ventilation releases during active mining	15.9	4532		
Releases from ore storage on surface	0.07	20		
Release from wastes on surface	0.66	188		
TOTAL RELEASES DURING ACTIVE MINING		4740		
Natural background emissions from surfaces affected by mining				
		4737		
NET INCREASE IN RADON EMISSIONS PER RRY	(182 tonnes)	3340		

SUMMARY OF RADON RELEASE FROM THE INACTIVE MINE

Upon shutdown of the mine, the ventilation system will be shut down, fans removed, and we assume that regulatory agencies will require the mine and ventilation shaft sealed so there will be no continuing radon release from vents. The ore stored on the surface will have been completely removed and only the accumulated waste rock may remain. The continuing radon release after shutdown of the mine will be just the quantity emitted by the waste stored on the surface, minus the natural background emissions which would have come from the area covered by the waste, summarized as follows:

Source	<u>Ci/Year</u>
Waste stored on surface	376
Natural background of area covered	<u>-6</u>
NET INCREASE IN RADON EMISSIONS AFTER SHUTDOWN DF MINE (following mining of 5700 tons $\rm U_3D_8)$	370
NET INCREASE IN RADON EMISSIONS PER RRY (182 metric tons)	13

SUMMARY AND CONCLUSIONS

Underground uranium mining, as determined from this study to date, representing measurements of radon release from several mine vents and from estimates of release from aboveground sources at one mine will produce a radon release of $16.6 \, \text{Ci/ton}$ of U_3O_8 mined. A small credit is included for a diminished background level at the waste storage site. At the end of the mine life, if the waste is left near the mine it will contribute about 370 Ci/yr indefinitely for the mine which has produced 5700 tons of U_3O_8 .

In terms of producing fuel for a year's operation of the reference reactor (1000 MWe LWR), this is equivalent to 3340 Ci/RRY (182 metric tons U_3O_8) during operation and 13 Ci/yr/RRY after shutdown, assuming 100% recovery of uranium from the ore. With 90.5% recovery of uranium from the ore, the respective values will be 3690 Ci/RRY and 14 Ci/yr/RRY.

The following characteristics were observed in the radon emissions from Mines 1 and 2:

- Very little short-term effect from mining operations other than ventilation practices;
- Large short-term variations in radon emissions from individual vents caused by changes in underground ventilation path;
- A distinct diurnal variation in response to barometric pressure changes;
- •Increased radon emission as the area ventilated increased;
- Radon emission closely related to the cumulative tons of ore extracted from two mines--less closely related to cumulative tons of U_3O_8 ;
- Relative standard deviation of radon concentrations for consecutive 4-hour integrated samples was $\pm 30\%$ for nine vents observed for about one month;
- •Concentrations of consecutive 20-minute integrated samples varied by up to a factor of 2 during a three-day interval; and
- •Emission rates of radon from individual mine vents per cumulative tons of ore extracted from mined-out areas were apparently reduced by bulkheading and backfilling.

The possibility of using a "grab-sampling" procedure to evaluate radon emissions from a large fraction of the industry appears reasonable. Initial monitoring of about 30% of the U. S. production facilities has been made. The majority of the remaining facilities should be monitored. Only a limited number of detailed individual mine production data has been obtained to date. Such data will be essential in developing predictive models of radon emission from underground mines.

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TABLE I

PRECISION OF 4-HOUR INTEGRATED SAMPLES

MINE - VENT	RELATIVE STANDARD DEVIATION (%)
1 - 1	9
1 - 2	14
1 - 3	13
1 - 4	18
1 - 5	17
1 - Total	9
2 - 1	26
2 - 2	30
2 - 3	13
2 - 4	15
2 - Total	17

TABLE II

COMPARISON OF EMISSION RATES FOR MINES 1 AND 2

	Vent	Rn Concentration (nCi/m³)	Vent Flow (%/min)	²²² Rn Emission (Ci/day) ¹	U ₃ O ₈ Production (ton/day) ¹	$\frac{227}{\text{Ci/ton U}_3 \text{O}_8}$
Mine 1	1	1200	275,000	0.49	0.0	
	2	200	850,000	0.25	0.068	3.7
	3	750	2,260,000	2.42	0.50	4.9
	4	420	932,000	0.56	0.062	9.0
	5	18D	1,090,000	0.29	0.16	1.8
Total			5,407,000	4.0	0.79	5.1
Mine 2	1	22D0	509,000	1.58)))
	2	2700	2,140,000	8.39 9.97	0.18	55.0
	3	275	832,000	.33	0.080	4.1
	4	1370	2,940,000	5.78	0.89	6.3
Total			6,421,000	16.1	1.15	14.0
Mines 1 + 2				20.1	1.94	10.4

Notes

¹ Tabulated values calculated from unrounded data then rounded.

TABLE 111
RADON EMISSION MEASUREMENTS PROVIDED BY MINE OPERATORS IN 1976

Mine	No/Vent	²²² Rn Emission (Ci/day)	Ore Production (ton/day)	²²² Rn E (Ci/ton ore)	mission (Ci/ton U ₃ O ₈)*
1	4	4.3	473	.009	4.5
2	4	5.1	340	.015	7.5
3	10	13.1	360	.036	18.
4	6	12.2	229	.053	27.
5	11	12.1	604	.020	10.
6	5	4.3	951	.005	2.3
7	4	3.7	956	.004	1.9
TOTAL	44	54.8	3913	. 014	7.0

^{*}Ore grade not reported. Average grade of 0.2% $\mbox{U}_3\mbox{O}_8$ assumed.

TABLE IV RELATIONSHIP BETWEEN ORE AND U308 PRODUCTION AND RADON EMISSION

Mine	Range of ²²² Rn Conc. at Vents (nCi/m³)	Ventilation (%/min)	²²² Rn Emission (Ci/day)	Ore Production (tons/day)	U3O8 Production (ton/day) ³	²²² Rn Emission (Ci/ton ore)	222 Rn Emission (Ci/ton U_3O_3)
1	90-1400	4.4 x 10 ⁶	4.0	461	0.78	.009	5.1
2	200-3800	6.4 x 10 ⁶ (1)	16.0	222	1.15	.073	14.0
3	240-9100	5.6 x 10 ⁶ (1)	22.0	360	0.72	.061	31.0
4	200-21000	4.8×10^{6}	40.0	229	0.46	.175	87.0
5	540-7000	11.4 x 10 ⁶ (1)	~23.0 ⁽²⁾	604	1.21	∿.039	∿19.3
6	12-2400	8.1 x 10 ⁶ ⁽¹⁾	12.0	951	1.90	.013	6.1
7	51-1100	11.0 x 10 ⁶ (1)	12.0	956	1.91	.013	6.2
ALL M	TINES COM8INED AS A COMP	POSITE.	129.0	3783	8.13	.034	15.9
SAME 8	BUT OMITTING MINE 5		106.0	3179	6.92	.033	15.3

Notes

 $^{^{1}}$ Flow rates obtained from mine operators. 2 One of thirteen vents not sampled. Its emission was estimated using average concentration of radon from other vents. 3 The $\rm U_{3}O_{8}$ quantities for Mines 1 and 2 were provided by mine personnel. Remaining estimates were taken from data provided in 1976.

TABLE V
RELATIONSHIP BETWEEN RADON EMISSION RATES AND MINE PARAMETERS

ZZZPn Fmis		zzzRn Emission	Mine Surface Area	ea Cumulative		222Rn Emission (Ci/day)		
Mine Vent	Vent	(C1/day)	(M²)	Production Ore		Per cm² of surface area (x 10 ¹²)	Per Cumulative Ton Ore (x 10 ⁶)	Per Cumulative Ton U ₃ O ₃ (x 10 ³)
ı	1	0.49	18,200	42,000	91	* 0.27	11.7	5.4
	2	0.25	13,600	22,000	34	0.18	11.4	7.4
	3	2.42	113,000	210,000	410	0.21	11.5	5.9
	4	0.56	22,500	34,000	35	0.25	16.5	16.0
	5	0.29	25,000	28,000	51	0.12	10.4	5-7
	Total (Corr (4.01 Coeff)²with ²²² Rn E	192,300 missions .98	336,000 0.99	621 0.97	0.21	11.9	6.5
2	1 + 2	9.97		1,060,000	4900		9.4	2.0
	3	0.33		41,000	96		8.0	3.4
	4	5.78		155,000	790	•	37.3	7.3
	Total	16.1		1,256,000	5786		12.8	2.8
	(Corr C	o eff)⁴ with ²²²Rn E n	nissions	0.77	0.80	•		
Overal	ll Total	20.1		1,592,000	6407		12.6	3.1
(Corr	Coeff)2 wi	th ²²² Rn Emissions		0.82	0.84			

TABLE VI

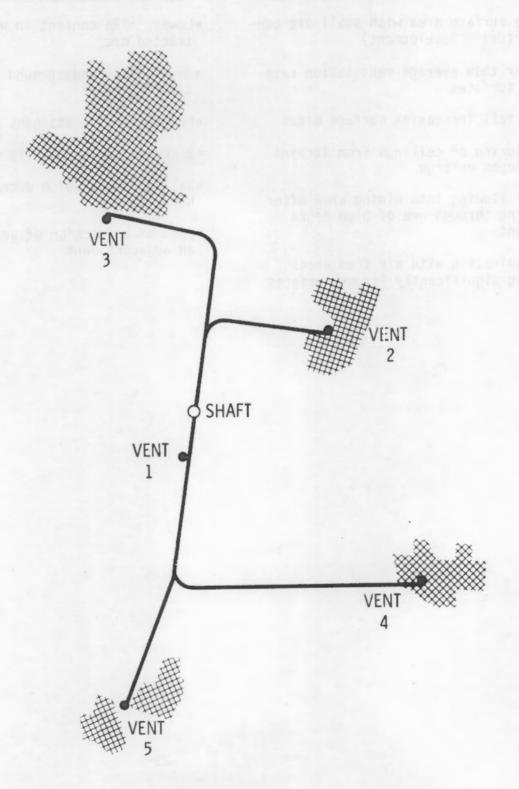
FACTORS CAUSING INCREASED RADON Ci/day PER CUMULATIVE TONS U308

- Large surface area with small ore content (Drift Development)
- Better than average ventilation rate past surfaces
- •Rock fall increasing surface areas
- Fracturing of ceilings from strains as stopes enlarge
- Water flowing into mining area after passing through ore of high ²²⁶Ra content
- Contamination with air from areas having significantly larger surfaces

FACTORS CAUSING DECREASED RADON per CUMULATIVE TONS U3D8

- Lower ²²⁶Ra content; in walls than in extracted ore
- Radon decay underground in stagnant ventilation areas
- Bulkheading to stagnant areas
- ·Backfilling, especially with barren sand
- •Wall coatings which decrease radon exhalation
- Loss of a fraction of ventilation air to an adjacent vent

FIGURE 1. Ventilated Areas of Mine 1



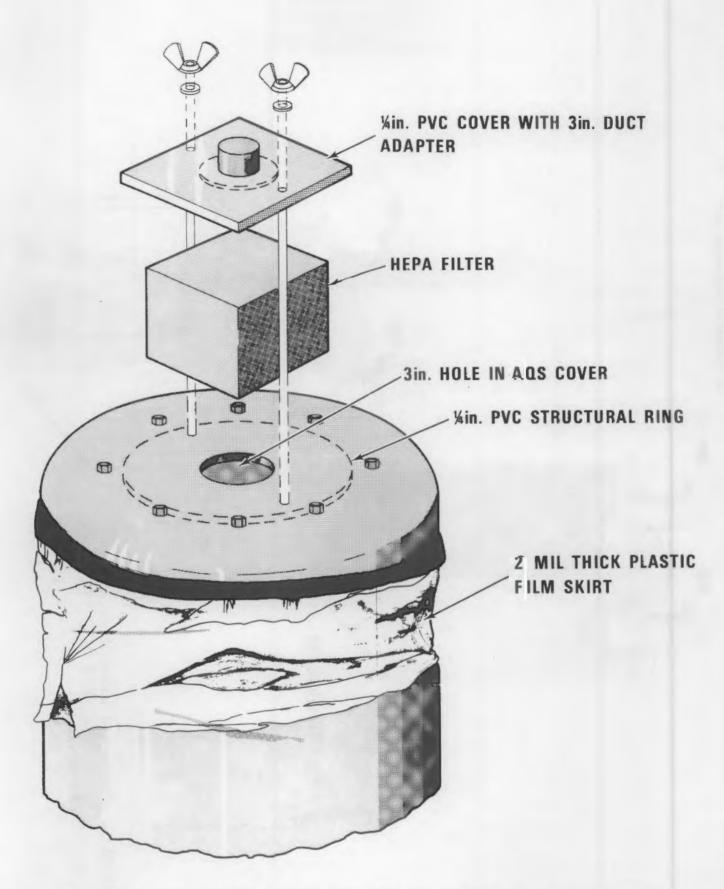


FIGURE 2. Modification for Conversion of the A.Q.S. Sequential Bag Sampler to Permit Vent Sampling

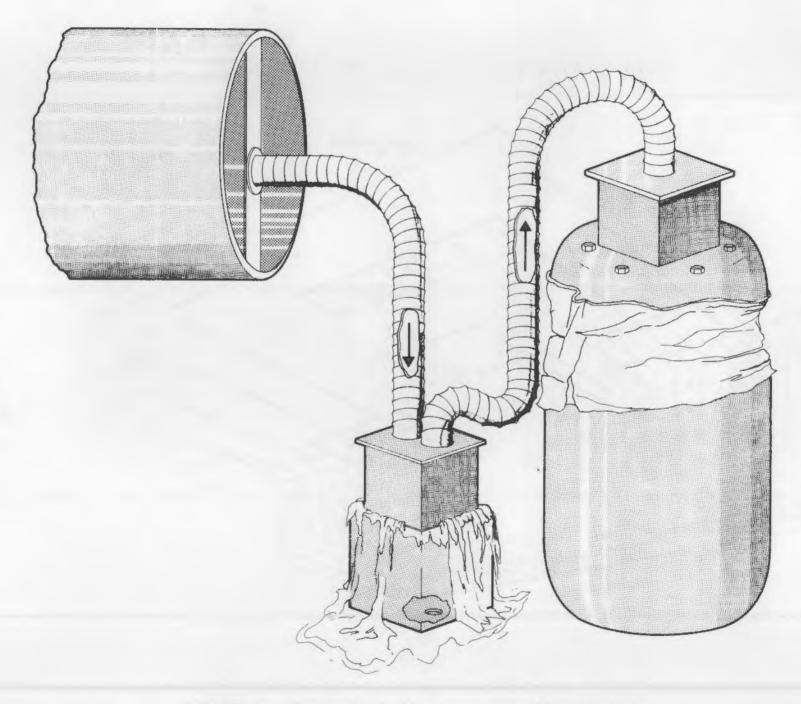


FIGURE 4. Sequential mine vent sampling system

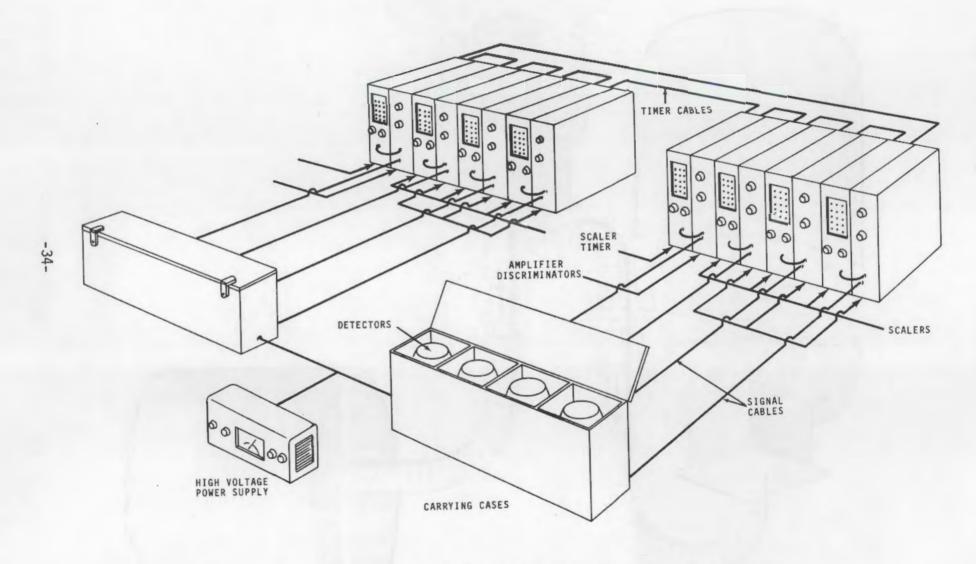


FIGURE 5. Diagram of radon flask counting system

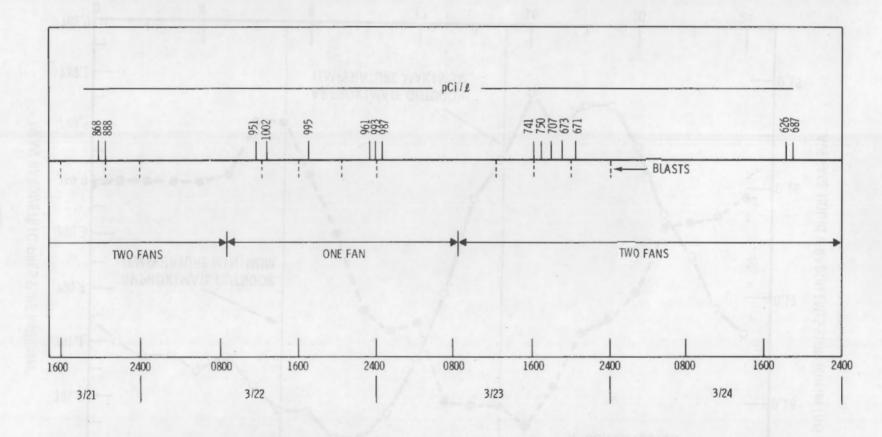


FIGURE 6. Comparison of ²²²Rn concentrations at Mine 1, Vent 3, with blasting schedules and the number of fans controlling the vent flow

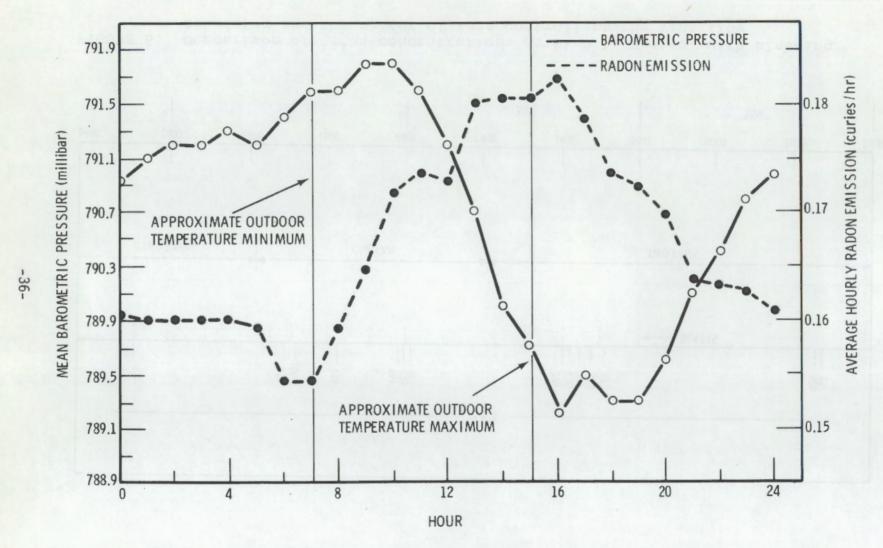


FIGURE 7. Comparison of the averaged ²²²Rn emissions at Mine 1 during August 1978 with the averaged barometric pressure readings

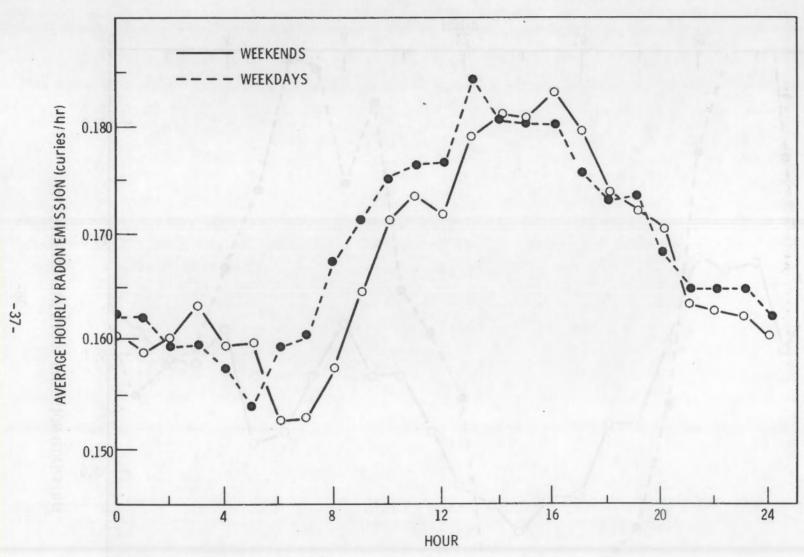


FIGURE 8. Comparison of the averaged $^{2\,2\,2}$ Rn emissions measured on weekdays and weekends at Mine l - August 1978

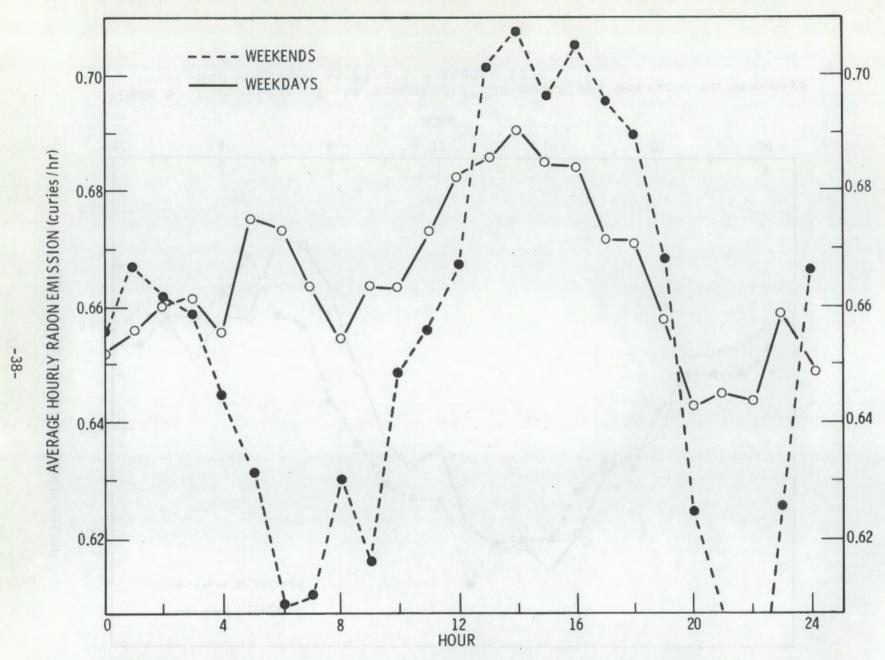


FIGURE 9. Comparison of the averaged $^{2\,2\,2}\text{Rn}$ emissions measured on weekdays and weekends at Mine 2 - September 1978

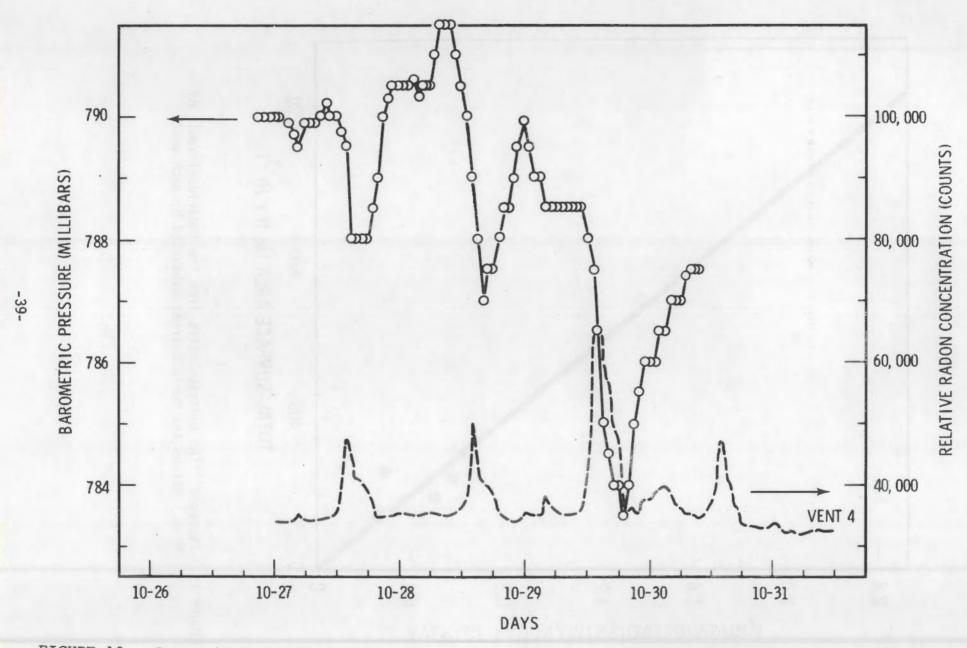


FIGURE 10. Comparison of consecutive 20-minute radon monitor counts with barometric pressure.

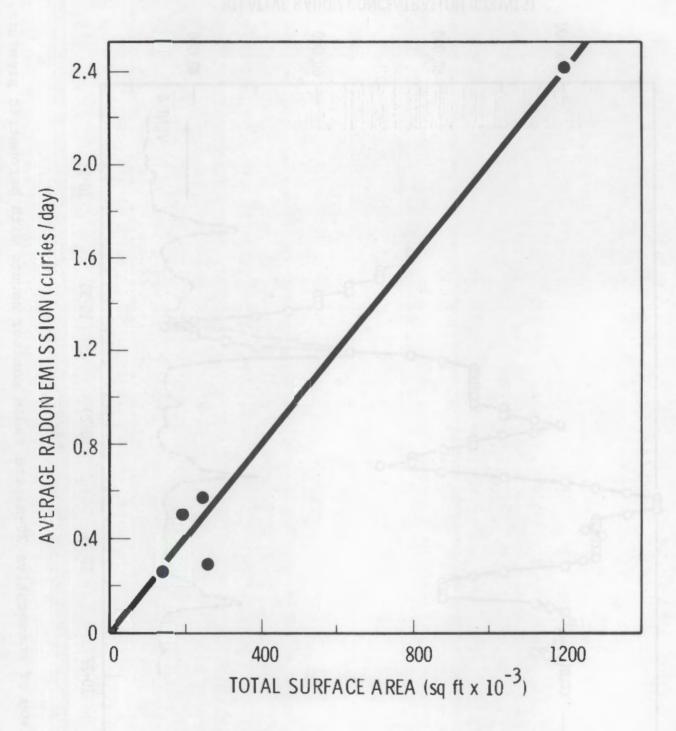


FIGURE 11. Averaged ²²²Rn emission rates from the individual vents of Mine I versus the surface areas exhausted by each vent.

APPENDIX A

CALIBRATIONS AND QUALITY ASSURANCE

Radon Calibrations Flask

Three commercial scintillation flasks and six of the 40 large shop-made flasks were taken to the Denver Federal Center for cross calibration. These units were evacuated and filled from a controlled atmosphere chamber designed by the Bureau of Mines. They were filled alternately with scintillation flasks which had been individually calibrated by personnel at the Center using ^{222}Rn swept from an NBS ^{226}Ra solution. After filling, the flasks being calibrated were transported to a temporary laboratory in the Grants, New Mexico area where the scintillation counting rates were measured. Meanwhile, the Bureau of Mines personnel measured scintillations from their calibrated flasks using their own instrumentation. After counting each flask several times, the pCi/\$\mathcal{L}\$ at the time of filling was calculated for the commercial flasks using the factory calibration of 5 c/m per pCi. The results are shown in Table A-1. Also shown for comparison is the average concentration measured with the Bureau of Mines' flasks during the sampling interval.

Table A-1

Radon-222 Measurements Using LAC-II Scintillation Flasks

Comparison of PNL Units Using 5 c/m per pCi
With Individually Calibrated Bureau of Mines Units

Instrument	Date Filled	Date Counted	222 Rn Concentration (nCi/m 3 ± Std. Deviation)
LAC II - 1207	7-28-78	8-2-78	1517 ± 17
U	7-28-78	8-3 - 78	1536 ± 22
Ω	7-28 - 78	8-4-78	1507 ± 25
LAC II - 1212	7-28-78	8-2-78	1495 ± 17
11	7-28-78	8-3-78	1514 ± 22
U	7-28-78	8-4-78	1520 ± 25
LAC II - 1205	7-28-78	8-2-78	1500 ± 17
н	7-28-78	8-3-78	1535 ± 26
n	7-28-78	8-4-78	<u>1517 ± 26</u>
	Average of P	NL Measurements:	1516 ± 7
	Average of B	ureau of Mines Measur	ements: 1526 ± 23

Since the agreement between these results was well within the precision of the measurements, the value 1516 nCi/m³ was used with the observed counting rates of the large shop-made flasks to determine their efficiencies. Eight photomultiplier (P.M.) tubes were used for these measurements. Each had slightly different detection efficiencies. Their relative efficiencies were determined on several occasions by recounting flasks on all P.M. tubes. Table A-2 shows the counting data from the large flasks filled at the Denver Federal Center after correction for radon decay and relative P.M. tube efficiency.

Table A-2
Initial Calibration Data for Large Shop-Made Scintillation Flasks

				(Net c	/min)		
Date Cou	unted	Flask 1	Flask 2	Flask 3	Flask 4	Flask 5	Flask 6
8-4-78	13:03	6175.1	5891.0	5759.4	6186.7	6144.8	5837.9
8-3-78	10:50	6103.3	5802.6	5758.7	6084.2	5827.0	5877.3
Average	=	6139.2	5846.8	5759.0	6135.5	5985.9	5857.6
	~	5954 ± 49		F054 ·	40		
Average	c/m/nCi/m³	± standar	d deviatio	on = 5954 ± 1516 ±	$\frac{49}{7} = 3.93$	± 0.04	

Also shown in Table II is the average counting efficiency for the large flasks calculated from the measured radon concentration.

Radon concentrations were determined from counting rates using the expression:

nCi/m³ = (c/m - Bkg c/m)(photomultiplier factor)⁻¹(3.93)⁻¹[
$$e^{\lambda(T_Ct^{-T}_S)}$$
]

where T_{ct} is the midpoint of the counting interval and T_{s} is the midpoint of the sampling interval.

Table A-3 shows the recalibration data for sixteen large scintillation flasks after the use of silicon optical compound was discontinued. This calibration was made in the field using the commercial scintillation flasks for cross-calibration. A commercial flask was filled after filling each group of four large flasks. All flasks were filled with an 18-min period to ensure sample homogeneity. The counting rates shown in Table A-3 were corrected for P.M. tube and decay to the filling time.

Table A-3
Recalibration of Shop-Made Flasks

Flask	Net c/m	Flask	Net c/m
Ε	11305	16	10481
AA	10307	Р	10865
1	10786	10	11075
С	10399	W	10393
LAC-II-1205	(2825 nCi/m³)	LAC-II-1212	(2817 nCi/m³)
8	11015	8	10286
12	10593	14	10691
S	10586	J	10635
I	10952	13	11017
LAC-II-1207	(2832 nCi/m³)	•	

Average c/m = 10712 ± 76

Average $c/m/nCi/m^3 = 3.792 \pm 0.028$

Precision of Measurements

Samples from all but four of the 73 vents measured to date contained sufficient radon to give in excess of 5000 counts during the counting interval. Counting statistics would predict a relative standard deviation of 2% or better for these data. However, examination of data from samples which had been given one or more recounts demonstrated that the precision was not as good as indicated by counting statistics. The cause of this variation was found to be the static charge induced on the exterior surfaces of the large flasks in handling. It was most pronounced when the relative humidity in the laboratory was low. The problem was partially corrected by spraying the exterior of the flasks with anti-static spray*. Three room-type humidifiers were also placed in the field laboratory. To determine the effect of static charge on prior measurements, 22 replicate grab samples which had been counted in the normal manner were recounted for four days. Flasks were either rubbed vigorously to induce a static charge or were sprayed with anti-static spray and counted at intervals. Finally they were sprayed and allowed to come to charge equilibrium for 18 hours in humid air and recounted. All counts were corrected to the fill time of the flasks. The counts were then normalized to the "equilibrium"

^{*}Van-Son Anti-Static Spray, Van Son Holland Corp. of America

discharged count for each flask. The average and standard deviation of the count ratios for normal, rubbed, and sprayed flasks were as follows:

Normal counting procedure	0.990 ± 0.089
Counts after rubbing flasks	1.008 ± 0.059
Counts after anti-static spray	0.770 ± 0.074

There was no significant bias in the averages of counts taken in the "normal" manner or after rubbing to induce a static charge. However, for several hours after spraying with anti-static compound, the counts were significantly low. To evaluate the effect of surface charge on the counting precision, the standard deviation for each class prior to normalizing to the "equilibrium" counts was calculated. Assuming that the actual radon concentration was constant while the samples were collected, these estimates reflect the variability of individual counts with our procedures. The relative standard deviation of the "normally" handled group was ±7.6%. After antistatic spraying and charge equilibration, the relative standard deviation was $\pm 3.8\%$. The former precision reflects the limiting precision of measurements taken prior to attempts to control static charge. The latter is more typical of the limiting precision of individual calibration measurements when longer equilibration periods were used after filling. For samples taken after the anti-static treatment was initiated, the limiting precision would be intermedidate because of the shorter time interval between filling and counting. These sample results would also be unbiased because flasks were not normally used for a day after the treatment, and thereafter the anti-static coating was left undisturbed unless it became necessary to clean the flask.

The background counting rates for the scintillation flasks were measured at intervals during the study. The backgrounds were not constant, but depended upon the activity levels of the samples previously collected in each flask. Typically, backgrounds ranged from 0.1% to 0.2% of the previous count. This background occasionally persisted after more than one cycle of flushing and pumping, indicating that the radon had diffused into the plastic walls and/or the scintillation paint. Since the background was essentially negligible in comparison to the observed count for all but four vents, an average instrument background was used for most calculations. Since only six measurements had potential errors exceeding 2% from the residual background, the added effort of making individual background measurements prior to each

filling was not warranted. The maximum potential error from this source from the sampling order used was 6% for the lowest concentrations encountered.

Flow Measurements

A comparison was made of the pitot tube used in this study with a second pitot tube from the same manufacturer used by a mine operator. The flow rate at each traverse point of a vent was measured using each instrument. The ratio of the flow rate measurements averaged 0.989 ± 0.017 at one standard deviation. This comparison demonstrates the inherent precision of measurements for differential pressures in the range of 0.5 to 1.0 inches of water encountered at this vent. There was no significant bias between the units.

In practice, one set of flow rate measurements was made at each vent. To evaluate the possibility that diurnal flow rate variations were occurring in response to the change of air density, the pitot tube was fixed in place at the largest vent of Mine 1. Readings were taken at intervals for a period of six days. The resultant flow rates are shown in Table A-4. Diurnal flow variations were insignificant at this vent, although a significant diurnal ²²²Rn variation was observed there.

Table A-4
Flow Rates Measured at Mine 1, Vent 3

Date	Time Measured	Feet/min.
9 -1- 78	11:44	4360
n.	13:02	4310
15	14:04	4360
1+	15:22	4330
и	16:24	4310
9-2-78	09:30	4310
11	12:47	4330
n	13:47	4330
n	16:30	4330
9-3-78	09:00	4280
9-4-78	10:03	4330
D	21:15	4330
9-5-78	09:56	4360
н	16:20	4400
9-6-78	09:19	4310

(Maximum Reading)/(Minimum Reading) = 4400/4280 = 1.03

The accuracy of the vent flow measurements is a function of the magnitude of readings taken and the uniformity of the air velocity within the vent. At one vent, the differential pressure seen by the pitot tube fell below the minimum readability of the manometer (approximately 0.01 in. to 0.02 in. H_20). These low values were encountered because of extremely low and non-uniform flow. A large diameter vent had been capped with a small diameter fan exhausting a hole in the center of the cap. The available pitot tube ports were in the large diameter vent pipe close to the fan. In this case, the relative standard deviation of the individual readings in the eight point traverse was 0.70. The relative standard deviation of the individual traverse measurements ranged from 0.10 to 0.23 at the other vents studied in detail.

Other than Mines 1-8 and Mines 11 and 12, the vent pipe configurations were not amenable to pitot tube flow rate measurements. At Mine 14 an attempt was made to place the pitot tube into the mouths of the vents to measure the flow in a four point traverse. These measurements must be considered tentative at this time. We are planning to remeasure vent flows with totalizing vane anemometers where appropriate in the future.

Diffusion/Leakage from Tedlar Bags

To ensure that radon was not escaping from the tedlar bags between sampling and transfer, three bags were filled from the test atmosphere chamber at the Denver Federal Center and transported to the field laboratory. After 10 days the air in the bags was transferred to small scintillation flasks and the radon measured. Two of the bags had been sealed by pinching the attached clear PVC tubing at a distance of about 4 cm from their inlet connectors. The average loss from these bags was equivalent to an escape half-time of 88 days. That rate of loss would cause a loss of 0.8% in the maximum 24 hours during which samples are contained in the bags. The third bag was purposely attached to a longer length of PVC tubing clamped at a distance of 30 cm from the connector. Loss from that bag was equivalent to a half-time of 20 days. Since the exposed portions of connecting tubing used in the sequential sampler was about 6 cm long, the diffusional loss was neglected.

APPENDIX B

TABULATION OF INDIVIDUAL GRAB SAMPLING MEASUREMENTS

M1ne	/Vent	Samp <u>Date</u>	ling <u>Time</u>	Stack Flow (1/mjn)	222Rn Concentration (nCi/m³)	²²² Rn Emission Rate <u>(Ci/hr)</u>
3	1	10/28	1003	454,000	240	.0066
	1	10/28	1002	454,000	290	.0079
	1	10/29	1249	454.000	342	.0093
	1	10/29	1250	454,000	· 368	.0100
	2	9/24	1006	406,000	7,600	.185
	2	9/24	1007	406,000	7,530	.183
	2	10/28	0956	406,000	7,870	.192
	2	10/28	0955	406,000	7,840	.191
	3	9/24	1027	304,000	7,440	.136
	3	9/24	1028	304,000	7,680	.140
	3	10/26	1637	304,000	3,560	.0650
	3	10/26	1636	304,000	3,380	0617
	4	9/25	1042	393,000	6,460	.152
	4	9/25	1043	393,000	5,620	.132
	4	10/26	1649	393,000	6,330	.149
	4	10/26	1650	393,000	6,370	.150
	7	9/23	1117	513,000	9.060	. 279
	7	9/23	1118	513,000	8,270	. 254
	7	10/26	1623	513,000	5,810	.179
	7	10/26	1624	513,000	5,560	.171
	8	9/24	1059	429,000	2,610	. 0673
	8	9/24	1100	429,000	2,450	.0632
	8	10/29	1322	429,000	8,310	.214
	8	10/29	1324	429,000	8,070	.208
	9	9/24	1045	889,000	1,260	. 0669
	9	9/24	1046	889,000	1,230	.0658
	9	10/28	1017	889,000	1,220	.0651
	9	10/28	1014	889,000	1,300	.0693
	10	9/25	1036	811,000	682	.0332
	10	9/25	1037	811,000	627	.0305
	10	10/29	1315	811,000	836	.0407
	10	10/29	1314	811,000	905	.0440
	11	9/24	1039	628,000	290	.0109
	11	9/24	1040	628,000	262	-, 0099
	11	10/28	1011	628,000	257	. 0097
	11	10/28	1012	628,000	293	.0111
	11	10/29	1301	628,000	367	.0139
	11	10/29	1302	628,000	341	.0129
	12	9/25	1026	778,000	307	.0143
	12	9/25	1027	778,000	251	.0117

					222gn	^{2 7 2} Rn	
Mine/Vent		Samp <u>Date</u>	ling <u>Time</u>	Stack Flow (%/min)	Concentration (nCi/m ¹)	Emission Rate (Ci/hr)	
4	2	9/22	1135	707,000	4,750	. 201	
	2	9/22	1138	707,000	4,920	, 209	
	2	10/25	1447	707,000	5.969	.253	
	2	10/25	1448	707,000	5,580	.237	
	5	10/25	1505	408,000	11,700	.287	
	5	10/25	1504	408,000	11,700	.286	
	6	9/23	1102	351,000	17,100	. 359	
	6	9/23	1103	351,000	16,500	. 347	
	6	10/25	1458	351,000	21,300	.448	
	5	10/25	1500	351,000	21,400	.451	
	7	9/23	1036	572,000	7,200	.247	
	7	9/23	1037	572,000	6,510	.224	
	В	9/22	1145	1,030,000	6,920	.426	
	8	9/22	1147	1,030,000	5,410	.333	
	8	10/29	1410	1,030,000	7,190	.443	
	8	10/29	1411	1.030,000	7,630	.470	
	9	9/23	1055	162,000	261	,0026	
	9	9/23	1056	162,000	275	.0027	
	9	10/25	1452	162,000	751	.0073	
	9	10/25	1453	162,000	660	.0064	
	10	9/23	1043	811,000	1,520	.0742	
	10	9/23	1044	811,000	1,510	.0733	
	11	9/22	1154	1,200,000	202	.0145	
	11	9/22	1155	1,200,000	214	0154	
	11	10/29	1418	1,200,000	311	.0224	
	11	10/29	1417	1,200,000	327	.0236	

Mine/Vent	Samp Date	ling Time	Stack Flow _(1/min)	***ZRn Concentration (nC1/m*)	222Rn Emission Rate
5 1	9/28	0912	612,000		(Ci/hr)
1			612,000	1,670	.0613
i	9/28	0913		1,660	.0611
1	10/27	1449	612,000	1,740	.0639
	10/27	1447	612,000	1,850	.0680
2	9/27	1051	428,000	2,340	. 0601
2	9/27	1053	428,000	2,390	.0613
2	10/25	1339	428,000	2,450	.0629
2	10/25	1338	428,000	2,150	.0553
3	9/28	0922	289,000	6,200	. 107
3	9/28	0923	289,000	5,720	.0990
3	10/24	1641	289,000	6,910	.120
3	10/24	1642	289,000	7,010	. 121
3	10/29	1446	289,000	6,500	.112
3	10/29	1447	289,000	6,360	.110
4	9/28	0930	1,320,000	1,940	.154
4	9/28	0931	1,320,000	1,720	.137
4.	10/24	1425	1,190,000	2,450	. 175
4	10/24	1427	1,190,000	2.280	. 163
5	9/25	1059	351,000	4.020	.0848
5	9/25	1100	351,000	4,060	.0856
5	10/24	1730	351,000	5,170	.109
5	10/24	1732	351,000	5,330	.112
6	9/25	1107	1,620,000	955	.0928
6	9/25	1108	1,620,000	948	. 0922
6	10/24	1750	1,620,000	990	.0962
6	10/24	1751	1,620,000	972	.0945
7	9/27	1156	556,000	512	.0171
7	9/27	1157	556,000	507	.0169
7	10/24	1711	556,000	568	.0189
7	10/24	1712	556,000	530	.0177
8	9/27	0955	1,860,000	780	.0873
8	9/27	0956	1,860,000	744	.0832
8	10/26	1548	1,860,000	842	.0941
8	10/26	1551	1,860,000	931	.104
10	10/24	1742	960,000	1,960	.113
10	10/24	1743	960,000	1,810	.104
10	9/27	1059	960,000		.0654
10				1,140	
	9/27	1058 isured Vi	960,000	1,340	.0769
(12	: Or Filed	işüred Vi		1980 ± 1850	- 0643
	0/29	0000	1,203,000	1980 ± 1850	∿.064)
13	9/28	0855	1,620,000	519	.0505
13	9/28	0856	1,620,000	540	.0526
13	10/24	1416	1,620,000	573	.0558
13	10/24	1418	1,620,000	716	.0697
14	10/25	1357	567,000	1,960	.0668
14	10/25	1359	567,000	2.020	.0688
15	9/27	1032	1,360,000	7	.0005
15	9/27	1033	1,360,000	7	.0005

					^{2 2 2} Rn	2 2 2 Rn
Mine/Yent		Sampling <u>Date</u> <u>Time</u>		Stack Flow (1/min)	Concentration (nCi/m³)	Emission Rate (Ci/hr)
6	1	10/25	1433	1,910,000	1,620	.186
	1	10/25	1434	1,910,000	1,800	.206
	1	9/26	1038	1,540,000	1,210	.112
	1	9/26	1039	1,540,000	982	.0907
	3	9/26	1023	1,360,000	2,320	. 189
	3	9/26	1024	1,360,000	2,350	.192
	3	10/24	1649	1,360,000	1,550	,126
	3	10/24	1650	1,360,000	1,480	.120
	.4	9/26	1107	1,300,000	\$15	.0403
	4	9/26	1108	1,300,000	388	.0303
	4	10/27	1417	913,000	722	.0396
	4	10/27	1416	913,000	696	.0381
	5	9/26	1141	2,200,000	799	.105
	5	9/26	1142	2,200,000	759	.100
	5	10/27	1429	2,040,000	894	.110
	5	10/27	1430	2,040,000	869	.107
	6	9/26	1057	1,790,000	13	.0014
	6	9/26	1058	1.790.000	10	.0011

Mine/Vent		Sampling <u>Date Time</u>		Stack Flow (R/min)	Concentration (nCi/m²)	222Rn Emission Rate (Ci/hr)
7	1	9/20	1101	2,660,000	968	.154
	1	9/20	1102	2,660,000	999	.159
	1	10/17	1657	2,260,000	914	.124
	1	10/17	1658	2,260,000	967	.131
	2	9/20	1314	2,830,000	965	.164
	2	9/20	1102	2,830,000	963	.164
	2	10/27	1244	2,590,000	1030	.161
	2	10/27	1243	2,590,000	1080	.169
	3	9/20	1145	2,540,000	826	. 126
	3	9/20	1147	2,540,000	792	.121
	3	10/17	1816	2,700,000	861	-139
	3	10/17	1817	2,700,000	756	.122
	4	9/20	1133	1,130,000	52	.0035
	4	9/20	1135	1,130,000	51	.0034
	4	10/17	1751	1,210,000	52	.0038
	4	10/17	1752	1,210,000	70	.0051
	5	9/21	1136	1,270,000	538	. 0409
	5	9/21	1137	1,270,000	471	. 0358
	5	10/17	1711	1,600,000	592	.0570
	5	10/17	1712	1,600,000	586	. 0564
	6	9/21	1146	802,000	142	.0068
	6	9/21	1147	802,000	141	.0068

Mine/	<u>Yent</u>	Sampl Date	ing Time	Stack Flow (£/min)	222Rn Concentration (nCi/m³)	²¹² Rn Emission Rate (C1/hr)
8	1	9/21	1208	353,000	886	.0188
	1	9/21	1209	353,000	913	.0194
	2	9/21	7201	408,000	1,330	.0326
	2	9/21	1203	408,000	1,800	.0440
	2	10/27	1304	408,000	1,880	.0460
	2	10/27	1306	408,000	1,970	.0483
	4	9/22	1106	398,000	2,270	. 0542
	4	9/22	1108	398,000	2,010	.0480
	8	9/22	1116	1,150,000	1,330	. 0915
	8	9/22	1118	1,150,000	1,280	.0878
	8	10/25	1315	1,150,000	1,420	.0976
	8	10/25	1314	1.150.000	1,620	.112

Mine	<u>/Vent</u>	Samp <u>Date</u>	ling <u>Time</u>	Stack Flow (2/min)	222Rn Concentration (nCi/m²)	223Rn Emission Rate (C1/hr)
9	1	10/27	1118		1,620	
,	1	10/27	1117		1,780	
	1	10/28	0940		1,640	
	1	10/28	0939		1,470	
					-	
10	1	10/18	1417		217	
	1	10/18	1418		231	
	1	10/18	1419		231	
						•
11	1	10/17	1148	2,130,000	1,470	. 187
	1	10/17	1151	2,130,000	952	.121
	1	10/26	1352	2,130,000	1,460	.187
	1	10/26	1351	2,130,000	956	.122
	2	10/17	1156	1,880,000	881	.0992
	2	10/17	1158	1,880,000	853	.0960
	2	10/26	1403	1,880,000	686	.0773
	2	10/26	1405	1,880,000	608	. 0685
	4	10/17	1205	1,970,000	461	.0545
	4	10/17	1205	1,970,000	503	. 0594
	4	10/26	1417	1,970,000	452	. 0535
	4	10/26	1416	1,970,000	498	. 0589
12	1	10/17	1228		1,140	
	1	10/17	1229		1,070	
	2	10/17	1215	2,120,000	1,020	.129
	2	10/17	1217	2,120,000	1,010	.128
	2	10/26	1448	2,120,000	1,110	.141
	2	10/26	1449	2 120,000	1,030	.131
	3	10/17	1233	1,670,000	648	.0651
	3	10/17	1234	1,670,000	772	.0775
	3	10/25	1503	1,670 <u>,</u> 000	695	.0698
	3	10/26	1504	1,670,000	721	.0725
13	1	10/17	1254	••	613	
• •	1	10/17	1253	••	586	
	ì	10/27	1206		467	
	1	10/27	1207		485	
	2	10/17	1303		1,030	
	2	10/17	1305	-+	1,030	
	2	10/27	1218		1,090	
	2	10/27	1219		860	

Mine/Vent		Sampling Date Time		Stack Flow _(&/min)	222Rn Concentration (nCi/m³)	²²⁵ Rn Emission Rate <u>(Ci/hr)</u>
14	1	10/20	1317	511,000	309	.0095
	1	10/20	1318	511,000	207	.0063
	1	10/23	3327	511,000	326	.0100
	1	10/23	1328	511,000	276	.0084
	2	10/20	1332	685,000	440	.0181
	2	10/20	1333	685,000	540	.0222
	2	10/23	1336	685,000	520	.0214
	2	10/23	1337	685,000	452	.0186
	3	10/20	1345	631,000	510	.0193
	3	10/20	1346	631,000	609	.0231
	3	10/23	1346	631,000	635	.0241
	3	10/23	1347	631,000	683	.0258
	4	10/20	1413	853,000	1,680	. 0858
	4	10/20	1415	853,000	1,710	.3877
	4	10/23	1404	853,000	1,430	. 0733
	4	10/23	1405	853,000	1,530	.0782
	5	10/20	1421	418,000	801	. 0201
	5	10/20	1422	418,000	730	. 2183
	5	10/23	1412	418,000	6 50	.0163
	5	10/23	1411	418,000	624	.0157
	6	10/20	1400	510,000	1,100	. 0337
	6	10/20	143}	510.000	1,050	.0322
	6	10/23	1354	510,000	852	.0261
	6	10/23	1355	510,000	909	.0278
	9	10/20	1435	65,900	968	.0038
	9	16/20	1436	65,900	961	.0038
	9	10/23	1422	65,900	1,120	. 0044
	9	10/23	1423	65,900	1,090	.0043

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