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Abstract

Kenneth Ladrach THE OCCURRENCE OF RADON IN SOME NORTH CAROLINA GROUNDWATER SUPPLIES (Under the direction of Dr. James E. Watson)

Approximately one hundred small public groundwater supplies in North Carolina were sampled. Analyses for radon-222 were performed by two methods, emanation and a newer liquid scintillation counting (LSC) method. Two primary goals were involved in this work, (1) comparing the two analysis methods listed above and (2) testing for an association between radon concentration in groundwater and the geology of the sampled site. The data show statistically significant differences in radon concentrations measured by the two methods. In 75 percent of the cases the liquid scintillation result was lower, indicating the possible need for refinement of this technique. The precision of liquid scintillation results was tested by comparing dual samples from each site. A paired difference T-test on the dual LSC measurements indicates that the mean difference between dual LSC measurements is equal to zero. Forty three of fifty two differences are less than 10 percent different. The radon concentration data show in general, higher radon concentrations associated with granite and gneiss/schist rock formations over those in mafic and metavolcanic formations. Samples from the coastal plain area had the lowest radon concentrations measured.

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INTRODUCTION

Two primary objectives were addressed in the study presented in this technical report. The first question this report addresses is whether two methods used in the Radiological Hygiene Laboratory to measure radon concentrations in water samples provide comparable results. The standard procedure of emanating radon from a glass sample collection bubbler into an alpha scintillation cell for counting (Lucas 1957) has been used by a number of researchers in the past (APHA 1976, Lee 1979, Michel 1980, Mitsch 1982). Previous work by Radiological Hygiene students (Strain 1978, Mitsch 1982 and Hayes 1984) utilized this type of equipment and procedures for emanation analyses of groundwater samples from wells in the phosphate mining region of eastern North Carolina. For the current project the emanation apparatus and procedures were used to measure radon concentrations in well water samples collected statewide. However we wanted to implement an alternative analytical technique that would be reasonably accurate, reliable and less time intensive. The liquid scintillation counting method described by Prichard and Gesell (Prichard 1977) and subsequently used in a nationwide study (Horton 1983) seemed a good candidate. Sample collection for liquid scintillation counting analysis is easy to perform but analysis requires a liquid scintillation counter plus blank and standard activity vials. Since a programmable liquid scintillation counter is present in the Radiological Hygiene

Laboratory an appropriate counting regimen was all that was needed. A synopsis of the procedures used by the Environmental Protection Agency in their nationwide study of radon in drinking water (Horton 1983) was obtained from Larry Kanipe (personal communication, current address: Tennessee Valley Authority, Muscle Shoals, Alabama 35660). A good liquid scintillation counting procedure would provide an excellent alternative to emanation because more samples could be analyzed in a shorter period of time without requiring the presence of someone to operate the equipment. An important question is whether the liquid scintillation counting procedure performs accurately and reliably in comparison to the emanation procedure. This is the reason for comparison of emanation and liquid scintillation counting results in this report.

The second question this report is concerned with is the distribution of radon concentrations in groundwater as a function of different geological regions of the state. This technical report examines radon concentrations in water samples from well sites classified in five major geological groups across North Carolina. A statistical test for significant differences in radon concentrations between geological groups is performed. The longer range goal of this type of work is to be able to predict with confidence the concentration of radon to be expected in a given groundwater sample based on site geological characteristics and other well site parameters. A separate report published

by the Water Resources Research Institute (Loomis 1987) goes into more detailed analysis of the distribution of radon concentrations as a function of geology and other hydrologic parameters.

LITERATURE REVIEW

Analytical techniques

For years the standard analytical technique for the determination of radon concentration in water has been emanation of radon from the water into an evacuated scintillation cell for counting (APHA 1985). More recently, liquid scintillation counting (LSC) techniques have been used to measure radon concentrations. For example, the air in a cave in Japan was analyzed for radon by counting scintillation fluid after bubbling air through it in a scrubbing bottle (Amano 1985). In another study, thoron and radon gas bubbling from a hot spring was collected in a syringe, liquid scintillator was added and the mixture transferred to a vial for counting (Yoshikawa 1986). Two researchers used LSC in conjunction with other methods of analysis to study radon concentrations in groundwater (Ohnuma 1982, Oliveira de Sampa 1980). Radon concentrations measured by LSC were compared with concentrations measured using an ionization chamber (Ohnuma 1982). The coefficient of variation in LSC measurements was given as 4.9 percent and the correlation between the two methods was given as 0.966 (Ohnuma 1982). Oliveira de Sampa (1980) fabricated scintillation cells by internally lining the walls of Erlenmeyer flasks with silver activated zinc sulfide. Samples were then analyzed by emanation and LSC. Both methods were reported to yield high efficiency and

reproducible results. Only 0.06 percent of the radon in the sample was reported to be lost by retention in the emanation system and 95 percent of the radon in the sample was reported to remain dissolved in the liquid scintillation cocktail (Oliveira de Sampa 1980).

In a review of methods for radiological analyses of drinking water, Blanchard (1985) cites four investigators who have used LSC to determine radon concentrations (Noguchi 1964, Homma 1977, Prichard 1977 and Horton 1983). High volume extraction of radon from water followed by LSC was performed by Noguchi (1964) and Homma (1977). This technique was used to indirectly measure radium-226 in environmental samples. Simplified procedures have been used to directly measure radon in water collected in low volume samples using commercially available liquid scintillation counters (Prichard 1977 and Horton 1983). Broad spectrum energy windows were used by both investigators; however, different scintillation cocktails were employed. The precision of paired LSC measurements in the study by Horton (1983) was assessed by plotting the average range between paired measurements against average concentration. A linear fit to the data produced a slope of 0.054, indicating about 5 percent degree of precision over the range of concentrations measured. The accuracy of the LSC technique was checked through participation in an interlaboratory study at the University of Texas in Houston and a comparison study with the University of South Carolina Department of

Geology. EPA results in the interlaboratory study compared well with the known values of controls. For the comparison study with USC, a set of ten samples was analyzed by LSC (EPA-EERF lab) and by emanation (USC Geology lab). The correlation between the two sets of measurements was 0.998. The USC data were found to be about 10 percent lower than the EPA data as observed in a scatter plot of the two datasets. The LSC method used by the EPA (Horton 1983) was used in this study for comparison with the emanation method because of the ease of sampling and analysis as well as the previously demonstrated measurement capabilities of the technique.

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In the remainder of the literature review the hazard of radon exposure is described in terms of increased risk of lung cancer induction, and a relationship is presented between the hazard of indoor radon and the potential contribution of radon in groundwater to this hazard. Finally, the question of the influence of geology on the radon content of groundwater is explored.

Radon hazard

Radon gas and associated daughter products have been a concern for some time. On the average, radon daughters contribute the largest fraction of annual lung dose from all the sources of natural background radiation (NCRP #45 1975 & #77 1984), see table 1. In table 1 the category "inhaled radionuclides" refers primarily to inhaled radon daughters.

In order to attempt to quantify the hazard from inhala-

Radiation source	Dose equivalent
Cosmic radiation a	28
Cosmogenic radionuclides	1
External terrestrial ^b	26
Inhaled radionuclides	450 (3000)
Radionuclides in the body	24 (40) ^d
Rounded totals	500 (3000)

^a Allowing for 10% shielding by buildings.

^b Allowing for 20% shielding by buildings and 20% by the body.

^c Does not include thoron and its daughters. The modified value allows for indoor exposure to radon daughter inhalation and a change in quality factor from 10 to 20 for alpha radiation.

d Allows for a change in quality factor from 10 to 20 for alpha radiation.

Table 1. Summary of lung dose equivalents (in mrem/yr) from various sources of natural background radiation. Doses are to the bronchial epithelium. Values in parentheses are latest estimates. Adapted from NCRP #77, 1984.

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tion of radon daughters the National Council on Radiation Protection and Measurements studied the available data on the effects of radon inhalation experiments involving animals and data on the effects of radon inhalation among underground uranium mine workers. Their examination of data on effects of inhaling radon daughters found that animal study results parallel epidemiological studies of mine workers inhaling radon daughters. Several important points are made. First, very high cumulative exposures, over 1000 working level months (WLM), are less effective at lung cancer induction per WLM than are more moderate cumulative exposures. Secondly, the highest lung cancer risk coefficients for humans (50 x 10E -6 lung cancers/yr/WLM) were found among those exposed to radon daughters later in their lives (NCRP #78 1984). Finally, the regions of the human lung receiving the greatest absorbed dose from radon daughters are the basal cells of the epithelial tissue in the upper airways of the tracheobronchial tree. In fact human lung cancers do appear predominantly in the upper airways of this region (NCRP #78 1984).

NCRP report #78 adopts an average lung cancer risk coefficient of 10 x 10E -6 cancers/yr/person/WLM averaged over all age and exposure groups. Through a time integrating risk model NCRP converts this to a lifetime risk of about 1.5 x 10E -4 lung cancers per WLM averaged over all age and exposure groups. This is comparable to a range given by the International Commission on Radiological Protection of 1.5

to 4.5 x 10E -4 lung cancers per WLM (ICRP 1981). The model uses the average risk coefficient of 10 x 10E-6 cancers/yr/person/WLM as well as an exponential term to account for the decrease in cancer appearance rate due to cellular repair and cell death over time. NCRP #78 presents tabulated lifetime lung cancer risks for environmental levels of radon daughter exposure per pCi/m³ or per WLM per year for different ages of first exposure and different durations of exposure, see tables 2 and 3. The risks at the lower radon daughter levels found under environmental conditions have been extrapolated down from the risks obtained from the higher radon daughter levels of the uranium miner data. The NCRP justifies this extrapolation by taking the conservative position that lung cancer induction is a stochastic or non-threshold type of response and therefore there is some risk even at the lower doses delivered by environmental levels of radon daughters.

The tabulated risk coefficients can be used directly to determine the lifetime risk for developing lung cancer given age at first exposure and duration of exposure as in the following example (NCRP #78 1984). Let the average environmental exposure equal 0.2 WLM/yr (NCRP #78 1984). Using table 3 at a lifetime exposure duration beginning at age one, the lifetime lung cancer risk equals 9.1 x 10E-3 per WLM per year. The lifetime risk for developing lung cancer would be the product of the exposure level times the lifetime lung cancer risk per WLM per year or (0.2

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		Lifetime Lung Cancer Risk							Lung Cancers in
Exposure	Age at First Exposure								
	1	10	20	30	40	50	60	70	of 10. Letrone.
1 Year	2.5×10^{-8}	3.6×10^{-8}	5.0×10^{-8}	7.1×10^{-8}	8.3×10^{-8}	6.7×10^{-8}	4.8×10^{-8}	2.7×10^{-8}	0.0051
5 Years	1.3×10^{-1}	1.9×10^{-7}	2.7×10^{-7}	3.8×10^{-7}	4.0×10^{-7}	3.1×10^{-7}	2.1×10^{-7}	1.1×10^{-7}	0.026
10 Years	2.9×10^{-2}	4.2×10^{-7}	5.8×10^{-7}	8.1×10^{-7}	7.5×10^{-7}	5.6×10^{-7}	3.6×10^{-7}	1.5×10^{-1}	0.051
30 Years	1.3×10^{-4}	1.8×10^{-6}	2.1×10^{-6}	2.1×10^{-6}	1.6×10^{-6}	1.0×10^{-6}	4.8×10^{-7}	1.5×10^{-7}	0.14
Life	3.6×10^{-6}	3.5×10^{-6}	3.0×10^{-6}	2.5×10^{-6}	1.7×10^{-6}	1.0×10^{-6}	4.8×10^{-7}	1.5×10^{-7}	0.21

-Lifetime lung cancer risk under environmental conditions* per pCi 222Rn/m³. Lifetime risk as a function of age and duration of erroduce

Radon to radon daughter ratio Rn/RaA/RaB/RaC equal to 1/0.9/0.7/0.7; unattached RaA/RaB/RaC equal to 0.07.

* For a population with age characteristics equal to that in the whole United States in 1975.

Table 2. Lifetime lung cancer risks for various ages of first exposure and durations of exposure. Reproduced from NCRP # 78 1984.

Lifetime lung cancer risk under environmental conditions per	WLM per year.* Lifetime risk as	a function of age and duration of
--	---------------------------------	-----------------------------------

2.0		1.00		Lifetime Lung	Cancer Risk				1
Exposure	Age at First Exposure								
	1	10	20	30	40	50	60	70	of 10° Persons*
1 Year	6.4×10^{-5}	9.1×10^{-5}	1.3×10^{-4}	1.8×10^{-4}	2.1×10^{-4}	1.7×10^{-4}	1.3×10^{-4}	7.0×10^{-5}	13
5 Years	3.4×10^{-4}	5.0×10^{-4}	6.9×10^{-4}	9.8 × 10 ⁻⁴	1.0×10^{-3}	8.4 × 10 ⁻⁴	5.5 × 10 ⁻⁴	2.8×10^{-4}	66
10 Years	7.7×10^{-4}	1.1×10^{-3}	1.5×10^{-3}	2.1×10^{-3}	2.0×10^{-3}	1.4×10^{-3}	9.1×10^{-4}	3.8×10^{-4}	130
30 Years	3.4×10^{-3}	4.8×10^{-3}	5.5×10^{-3}	5.5×10^{-3}	4.2×10^{-3}	2.5×10^{-3}	1.3×10^{-3}	3.8×10^{-4}	380
Life	9.1×10^{-9}	9.1×10^{-3}	7.7×10^{-3}	7.7×10^{-3}	4.5×10^{-3}	2.7×10^{-3}	1.3×10^{-3}	3.8×10^{-4}	560

* For radon daughters measured under environmental rather than underground mining conditions.

* For a population with age characteristics equal to that in the whole United States in 1975.

Table 3. Lifetime lung cancer risks for various ages at first exposure and durations of exposure. Reproduced from NCRP # 78 1984. WLM/yr)(9.1 x 10E -3 per WLM per year) = 1.8 x 10E -3 lung cancers. This risk multiplied by the size of the U.S. population would yield the number of excess lung cancers to be expected in a lifetime due to exposure to average environmental levels of radon daughters. The 0.2 WLM/yr exposure can be expressed as continuous exposure to a radon concentration of 0.8 pCi/L by the conversion shown below (assuming 50 percent equilibrium of radon daughters).

$$\frac{0.2 \text{ WLM}}{\text{yr}} \quad \frac{\text{Yr}}{50 \text{ WM}} \quad \frac{200 \text{ pCi/L}}{\text{WL}} = 0.8 \text{ pCi/L}$$

By the same conversion process an exposure level of 1.0 WLM/yr corresponds to a concentration of 4 pCi/L which is the indoor radon concentration at which remedial action is recommended by the E.P.A., assuming 50% equilibrium of radon daughters (EPA 1986).

The source of airborne radon is radium-226 decay in the earth's crust (NCRP #45 1975, NCRP #77 1984, NCRP #78 1984). The chemically inert gas emanates from porous rocks and soils into the air above ground. Homes which are located on top of soils with high emanation rates are effective at trapping significant amounts of the emanating radon if their ventilation rates are low. Relatively high indoor radon concentrations (4 pCi/L or more) can be reduced roughly to the outdoor concentration by a ventilation rate of about four air changes per hour (NCRP #78 1984). The average outdoor radon concentration is often given as 0.2 pCi/L (NCRP #78 1984, EPA 1986). Most dwellings seldom attain the high ventilation rate necessary to ensure low indoor radon concentrations. As a result, houses in many areas of the United States have high radon concentrations indoors. It is estimated that about two percent or 1.0 to 1.6 million of the houses in the U.S. have indoor radon concentrations at or above 8 pCi/L, see figure 1 (Nero 1986). This is equivalent to about 2.0 WLM/yr which is the exposure limit recommended by the NCRP for an individual of the general population (NCRP # 77 1984). The exposure limit for a radiation worker is 4 WLM/yr (NCRP #77 1984).

Radon in groundwater

Although the greatest contribution to indoor radon is from soil emanation, radon will also emanate from building materials if significant amounts of uranium-238 decay products are present and from the home's water supply if it contains significant concentrations of radon. The latter possibility is of concern in this groundwater sampling project conducted in North Carolina.

A chemically unreactive gas is retained in water only to the degree that it is soluble in water. Radon is slightly soluble in water and can therefore be transported by water. But it can also easily escape from the water into the airspace above it if the water lies stagnant, or even more so if the water is agitated or aerated. The water supply to a house enters via a sealed system of plumbing that prevents escape of radon, but at points of direct water use there is usually considerable aeration and therefore





Figure 1. Distribution of radon concentrations in U.S. homes. Reproduced from Nero 1986.

opportunity for the radon to be released into the home environment. Bathroom, laundry and kitchen facilities are major sites where radon from the water supply could enter the home.

The questions of interest then become, how much radon is present in a typical water supply and how much radon is released indoors from a given level of radon in the water? These are difficult questions to answer accurately and require collecting a lot of data. It has been estimated that a radon concentration of 10,000 pCi/L in a home's water supply will contribute approximately 1 pCi/L radon indoors (Duncan 1976). This relationship is an average and will vary somewhat as characteristics vary from house to house in terms of their ability to trap the emanating radon.

In addressing the question of radon concentrations in water supplies the first consideration should be the source of the water. Dwellings that draw on open bodies of water such as lakes or rivers should not exhibit a high radon concentration in their water because most of the gas will be released through the large surface area available for emanation before the water enters the home. Homes supplied by large groundwater systems should also be less likely to show high radon concentrations in the water entering them because the size of the system usually results in long time periods between extraction of the water from the ground, and its use in individual homes. This time factor affords greater opportunity for natural decay of radon and radon

daughters and emanation of radon from the water while stored in tanks or towers. By contrast, in small groundwater supplies water is used in homes much sooner after removal from the ground. If the formations containing the groundwater are rich in the parent nuclides of radon and if the rock is porous enough radon can easily dissolve in the water and be transported to sites where the water is tapped for human use.

Geological influence

The question regarding how much radon is in a groundwater supply relates to the long range goal mentioned earlier, the formulation of a predictive model of radon concentration based on site characteristics and well parameters. In order to describe such a model different factors that influence radon concentrations need to be identified.

One factor is the type of rock from which the groundwater originates. Since radon is a link in the natural uranium decay series, water coming from rock formations rich in uranium-238 or its decay products is likely to contain a lot of radon. Previous studies have found significantly elevated radon concentrations in North Carolina groundwater supplies (Sasser and Watson 1978, Horton 1983) with one third of the samples in the former study showing radon concentations over 2000 pCi/L and several samples in the range of tens of thousands of pCi/L. A review of radon

highest concentrations to be in the Appalachian-Piedmont regions of the eastern states (Hess 1985) which includes a large portion of North Carolina. The Appalachian-Piedmont area is comprised of granite rock formations which are characteristically high in uranium-238 from which radon is formed.

Comparison of radon concentrations measured in North Carolina (measured in previous studies) with site geology showed radon concentration to be associated with rock type. The highest concentrations were found in groundwater from sites located in granite formations (geometric mean equals 5900 pCi/L, Loomis 1985). Gneiss, schist and metavolcanic regions of North Carolina contained groundwater with lower radon concentrations with a geometric mean of about 1200 to 1300 pCi/L and the rock types of the coastal plain area showed the lowest radon concentrations in groundwater sampled (Loomis 1985). The association between site geology and radon concentration in groundwater is examined as one of the two objectives of this report. A statistical test for differences in measured radon concentrations between the geological groups sampled in North Carolina is applied in the data analysis section of this report.

Other factors besides geology could influence variations in radon concentrations in groundwater. Among them are the porosity of rock strata, the ratios of parent nuclides of radon in the rock, the quantity of water present in the rock, the rate at which the water is pumped from the

ground and time or climatic variations. Some of these are addressed in the report to the Water Resources Research Institute (Loomis 1987) but are not given any more attention here.

For this study it was decided that small public water supplies would be used for sample collection. In North Carolina these are systems serving 25 persons or more with water from one to several wells located in the vicinity of the user population. An advantage of sampling public water supplies is that the state requires that public records be kept for each well in such a supply. This meant that owner contact information and information needed to locate potential sampling sites was on file. These are important items of information to have in conducting a statewide sampling project of this size. Also available on file were the driller's pumping tests. These were used to obtain information about each well for calculation of hydrologic parameters under consideration for the predictive model of radon concentration.

MATERIALS AND METHODS

Two hundred and nineteen wells were selected for potential sampling. The goal was to actually sample 100 of them for the final dataset (20 from 5 geological groups). All sites were wells that serve as small public water supplies. Although it was not a requirement that they were actually being used at the time of sampling, this was usually the case. Choosing from public well water supplies facilitated gathering owner contact information, pumping capacity data and geological information for each site from public records.

Data on individual wells needed for contacting the owner and locating the well were obtained by Dana Loomis from the state's Division of Health Services Water Supply Branch computer files. The owners were initially contacted by mail describing the sampling project and soliciting permission to collect a sample from their well. A copy of the driller's well record was also obtained from the Ground Water Section of the North Carolina Department of Natural Resources and Community Development for each well selected. These provided data on exact location, total depth, drilling date and the driller's pumping test. Pump test data were used to calculate values of hydrological parameters for each well.

Each site was assigned to one of five generalized geological categories or rock types. This was based on surface geology at the site as determined from U.S. Geological Survey maps. The final dataset was comprised of wells sampled from granite rocks, gneiss and schist rocks, metavolcanic rocks, mafic rocks and the rocks of the coastal plain region of North Carolina.

From the selected sites a sample of 100 was randomly chosen (20 in each of the 5 rock groups). Their locations were marked on a state map to organize sampling trips that would maximize collection efficiency. Owners of wells were contacted by telephone to arrange specific meeting times for sampling each well.

Two sample analysis techniques were used during this work. The technique of radon emanation into a scintillation cell was performed on all samples, and a liquid scintillation counting procedure was performed concurrently on about half of the sampled sites (Horton 1983). The emanation process will be discussed first.

Emanation

Sample collection for analysis using the emanation technique was made at a water faucet located as close to the wellhead as possible. Every attempt was made to obtain samples that were representative of fresh groundwater. This meant avoiding water from pressure tanks, storage tanks, water towers or outlets distant from the wellhead. In addition the pump in the well was switched on and the collection valve was opened to allow water to flow from the well for several minutes before sample collection. To collect the sample the flow rate was greatly reduced and a

rubber hose was fitted to the faucet. The other end of the hose was attached at the collection stopcock S3 of the emanation bubbler shown in figure 2. Water was allowed to enter the bubbler displacing the air through the open stopcock at the top of the bubbler (stopcock S1). Stopcock S2 remained closed. If turbulence was evident the water was discarded and the flow rate reduced further until a sample was obtained without aeration. Since radon is a chemically unreactive gas it may be lost rather easily from the sample if there is much aeration of the water during collection. When the bubbler was approximately three quarters full collection was stopped by rapidly closing stopcocks S3 and S1 in that order. The bubbler was then disconnected, labelled and carefully stored for transportation. Time and date of collection were noted for each sample. One sample for emanation analysis was taken at each site. Samples were transported back to the Radiological Hygiene Laboratory for analysis as soon as possible. Samples were emanated within one to two days of the time of collection.

The emanation apparatus used for analysis is shown in figure 3. It consists of stainless steel seamless tubing (I.D. of 0.25 inches), clear plastic tubing and hose clamps of various sizes, glass stopcocks, silicone sealant, Whitey valves, Swagelok fittings, calcium sulfate desiccating material and a Fisher vacuum gauge. Prior to use each day the system was evacuated in order to assess its ability to maintain sufficient vacuum. The loss of a maximum of 3



(Reproduced from Mitsch 1982)



inches mercury over thirty minutes was judged as acceptable. The scintillation cell to be used for the emanation of a given sample was flushed and filled several times with helium gas and counted for background for thirty minutes just prior to the emanation. Helium was used during background counting because helium was also used during the The emanation was performed following the emanation. procedure outlined in the technical report of Barry Mitsch (Mitsch 1982). The sample bubbler and scintillation cell (with all stopcocks closed) were attached to the emanation system as shown in figure 3. The helium supply was then attached, valves V1 through V3 were opened, stopcock S4 was closed and the system was evacuated by switching on a vacuum pump attached as indicated in the figure. Stopcock S5 on the scintillation cell was then opened and the drop in vacuum was noted to be certain the cell was evacuated. After 1 to 2 minutes the system was sealed off from the pump by closing valve V2. The vacuum pump was returned to atmospheric pressure by opening stopcock S4. The emanation was initiated by very slightly opening stopcock S1 at the top of the bubbler. The difference in pressure draws gases out of the water and into the system. Once the pressure equalized the stopcock was opened fully. The bubbling was maintained by slightly opening stopcock S2 on the bubbler to allow pressurized helium to continually flow through the bubbler and the system. The rate of helium flow was used to govern the bubbling for the remainder of the emanation.

During the emanation, radon and helium gas pass through filter paper barriers and desiccating material where water vapor and radon progeny attached to particulates become trapped. Care must be taken not to let water rise up into the system during emanation or the desiccant and filters are rendered useless. After a thirty minute bubbling time the process was stopped by closing stopcock S5 on the scintillation cell sealing in any radon that was transferred from the sample. Next, stopcocks S1 and S2 were closed to seal the water in the bubbler. The rest of the system was returned to room pressure and the cell was disconnected and stored in a light tight box to allow equilibration between radon and radon progeny. A minimum equilibration time of four hours was always used.

The electronic counting system used is depicted in figure 4. The components used are indicated. For a complete description of equipment settings used see appendix I. Prior to any background or sample counting for the day and at the end of counting the consistency of the system was tested. A standard activity scintillation cell was used for this testing. Ten one minute counts were recorded and averaged. For background and sample counting, scintillation cells were counted for thirty minutes. Since emanation was performed for thirty minutes, a series of several samples could be efficiently analyzed by simultaneously emanating one sample into a scintillation cell while counting the background in the next cell. Each sample scintillation cell



Figure 4. Block diagram of electronics system used for counting scintillation cells. Cell was placed inside light shield on top of photomultiplier tube. For a complete description of settings, see appendix I. Adapted from Mitsch 1982.

was counted once after a minimum four hour ingrowth. Liquid scintillation

Sample collection for liquid scintilltion analysis was made immediately after and at the same outlet where samples were taken for emanation analysis. To collect the sample the water flow rate was reduced, clear plastic tubing was fitted to the outlet and a four inch diameter funnel was attached to the free end of the tube. Holding the funnel upright the water was allowed to pool and slowly overflow the edges until all air pockets had been purged from the tubing and no turbulence was evident in the water. A 20 cc syringe with 18 gauge needle was inserted one to two inches below the water surface. As the water continued to flow the syringe was flushed a few times discarding the water each time. The final 10 ml sample was drawn into the syringe and immediately expelled at the bottom of a 20 ml glass liquid scintillation counting vial already containing 10 mls of a mineral oil based scintillation fluid. Care was taken to draw the water into and expel it from the syringe slowly to minimize aeration of the sample. The dense water layer remains at the bottom of the vial and the organic layer on top helps prevent the escape of radon as the water is introduced into the vial. Two vials were filled at each site and the time and date of collection were noted. Once transported to the Radiological Hygiene Laboratory the vials were counted within one to two days of the time of collection.

Counting was done using a Packard Tri-Carb 300 liquid scintillation counter preprogrammed to count each vial for 50 minutes or until the percent deviation was down to 2 percent. The energy windows were set to count from 0 to 2000 KeV. Two background vials were counted with each batch One background vial contained only 10 ml of of samples. scintillation fluid and the other contained scintillation fluid plus 10 ml of distilled water. No standard activity vials were counted with samples because none were available when sampling for liquid scintillation counting began. Just prior to starting counting, each vial was shaken for about 15 seconds to mix the two fluid phases. According to the EPA protocol (Horton 1983) the radon in the water sample preferentially dissolves in the organic scintillator layer and any radium present remains dissolved in the aqueous layer. The batches of vials were counted after four hours in order to allow for equilibration of radon daughters. See appendix II for step by step details of the liquid scintillation counting procedure used.

Calibrations

In order to calculate sample radon concentrations from count rate data collected by either analysis technique a calibration factor was needed relating count rate to activity present in the sample. This was obtained by analyzing a sample of known activity in exactly the same way that unknown samples were analyzed. The net count rate observed was then divided by the known activity contained in

the standard. This factor (counts per minute per unit activity) was then divided into the count rate from an unknown sample to convert to sample activity.

Separate standard activity sources were needed for the two analysis procedures. Two standard bubblers made previously by a Radiological Hygiene student were initially used for the emanation system (Carver 1980). By allowing the source to remain sealed for approximately thirty days the activity of radon equilibrates with that of the radium. The bubbler is then processed as if it were a sample of unknown activity. Calibration factors obtained from analyses on the two standard bubblers were inconsistent with values obtained in previous work by Barry Mitsch (Mitsch 1982).

Since the completion of sampling, two other sources of radium have been obtained. The first was through participation in an interlaboratory quality control program with Lockheed Engineering and Management Services, Las Vegas, Nevada. The program was designed to assess the reliability of environmental radon measurement efforts by laboratories across the country and to test the performance of a new radium-226 source package they had fabricated. We were provided with a bottle containing a known amount of radium-226 activity dried on a piece of filter paper which was sealed between two pieces of clear plastic. The sealed source packet was immersed in the bottle full of water. Radon generated from the radium diffuses through the plastic seal into the water while the radium does not (as long as the water tight plastic seal remains intact). By allowing the bottle to remain sealed for thirty days the radon activity equilibrates with the radium activity. The standard activity water was analyzed by carefully transferring aliquots to bubblers and counting vials. The procedure can be repeated by simply refilling the source bottle and allowing for ingrowth again.

The second new radium source was obtained from the Environmental Protection Agency in Las Vegas, Nevada in the form of an aqueous solution. From this solution dilutions were made in two bubblers and five sealed liquid scintillation counting vials. See appendix III for a step by step description of the dilution procedure used to make these standards. Calibration factors obtained from analyses of these dilutions were used for final calculation of radon concentrations.

DATA ANALYSIS

One hundred sampled wells are included in the final dataset. All wells were sampled for emanation analysis. Only fifty two were sampled for liquid scintillation analysis because sampling began before the liquid scintillation counting procedure had been set up. Samples were collected from May through November 1986. The radon concentration data obtained for the 100 sites are shown in appendix IV. The data are sorted by five geological classifications of sites sampled. Radon concentrations in pCi/L were calculated from net count rates obtained from sample analyses. The relationship used is shown below,

Rn-222 conc. (pCi/L) = $\frac{X}{Y}$ $\frac{1000 \text{ ml/L}}{V}$ exp (λt)

where;

X = net count rate (cpm)

Y = calibration factor (cpm/pCi)

V = sample volume (ml)

) = radiological decay constant for radon (1.8 x 10E-1 daysE-1)

t = elapsed time from sample collection to midpoint
of count (days).

Calibration factors

As stated in the methods section, samples of known activity were analyzed to obtain calibration factors in counts per minute per picocurie (cpm/pCi). Calibrations for the emanation system, obtained from analyses of two existing emanation standards were inconsistent. One value (4.73 cpm/pCi) agreed well with previous results while the other did not (3.27 cpm/pCi). It was concluded that these old emanation standards were no longer reliable, and that an average calibration factor from previous work (4.61 cpm/pCi) should be assumed (Mitsch 1982) until the old standards could be replaced.

A new sealed radium-226 source from an interlaboratory quality control study provided a convenient means of generating water containing a known radon activity. Aliquots of this water were dispensed into emanation bubblers and liquid scintillation counting vials and analyzed in the usual manner. Results of this work are shown in table 4. Results from other laboratories participating in the study are shown in table 5.

In addition, an aqueous radium-226 source obtained from the Environmental Protection Agency in Las Vegas was diluted into a stock solution which was used to make sealed standard bubblers and vials. The procedure used for this is given in detail in appendix III. Results from analyses of these new standards are shown in table 6. The emanation calibration factor in table 5 (4.89 cpm/pCi) agrees well with values obtained in earlier work (Mitsch 1982). However, this value and the liquid scintillation calibration from table 5 were not used to calculate radon concentrations because of the potential for loss of radon during handling of the standard activity water. The average calibrations listed in table 6
analytical method	source activity (pCi)	calibration #1 (cpm/pCi)	calibration #2 (cpm/pCi)	calibration #3 (cpm/pCi)	average calibration (cpm/pCi)
Lucas cell emanation	832	4.84 ± 0.039	4.93 <u>+</u> 0.039	4.91 <u>+</u> 0.039	4.89 <u>+</u> 0.039
liquid scintillation	832	8.68 <u>+</u> 0.097	8.62 <u>+</u> 0.096	7.60 <u>+</u> 0.087	8.30 ± 0.093

Table 4. Calibration factors obtained with radium-226 source from interlaboratory study.

Emanation method

Laboratory	Calibration factor
	(cpm/pCi)
5	3.79 + 0.08
8	4.24 + 0.04
14	2.50 ± 0.04
16	4.78 + 0.13
*17	4.89 ± 0.04
19	4.67 ± 0.21
21	4.66 ± 0.15
24	4.57 ± 0.24
25	1.53 + 0.13
28	0.105

Liquid scintillation method

	(cpm/pCi)	
1	8.88 + 0.00	
2	8.25 ± 0.23	
3	7.26 ± 0.19	
4	8.45 ± 0.05	
6	8.13 ± 0.34	
7	7.50 + 0.23	
10	8.59 + 0.06	
11	7.51 ± 0.45	
13	6.75 ± 0.08	
15	7.70 ± 0.17	
*17	8.30 ± 0.49	
18	8.40 + 0.08	
18	8.12 + 0.37	
20	8.08 + 0.10	
21	8.19 + 0.05	
21	9.12 + 0.09	
23	6.82 + 0.003	
26	9.00 + 0.09	
27	3.64	
27	3.34 ± 0.47	

* indicates Radiological Hygiene Lab

Table 5. Calibration factors obtained by participants in interlaboratory study. Adapted from personal communication from E.L. Whittaker, Lockheed Engineering and Management Services Company, Environmental Programs Office, 1050 E. Flamingo Road, Suite 120, Las Vegas, Nevada 89119.

Lucas Cell Emanation

Activity (pCi)	Calibration (cpm/pCi)
238	5.67 <u>+</u> 0.032
476	5.38 ± 0.022
	* 5.53 ± 0.027 (average)

Liquid Scintillation Counting

Activity (pCi)	Calibration (cpm/pCi)
238	10.87 ± 0.11
476	10.96 <u>+</u> 0.11
714	10.85 <u>+</u> 0.11
952	10.78 <u>+</u> 0.11
1190	10.84 ± 0.11

10.86 ± 0.11 (average)

- Table 6. Calibration factors obtained with dilutions from aqueous radium-226 source from the Environmental Protection Agency in Las Vegas, Nevada.
 - * Analyses of the two standard activity emanation bubblers has been repeated. The resulting calibrations were 5.61 cpm/pCi for the 238 pCi standard and 5.11 cpm/pCi for the 476 pCi standard. The overall average is 5.44 cpm/pCi or 81 percent efficiency.

for emanation and liquid scintillation counting were used for final calculations of radon concentration. The standards from which these data were obtained are sealed and do not require handling of an aliquot prior to analysis.

The final average calibration factor used to calculate the emanation radon concentrations (table 6) represents a 20% increase over the average value of 4.61 cpm/pCi previously used (Mitsch 1982). Although this is a significant increase, it should be noted that quality assurance emanation experiments performed at U.N.C. in cooperation with the E.P.A. in Las Vegas produced consistently high radon measurements (Mitsch 1982). It was proposed that error in the calibration factor was the greatest contributor to these high radon measurements (Hayes Consistently high radon measurements could be 1984). explained by the calibration factor being too low (see previous expression for calculating radon concentration). The quality assurance radon measurements performed by Mitsch were an average of 23 percent too high.

Exploratory data analysis

The data in appendix IV include radon concentrations determined by both emanation and liquid scintillation analysis for fifty two out of one hundred sites sampled. An objective of this study was to determine whether the liquid scintillation counting procedure produced results consistent with those of the standard emanation method. An exploratory examination of the data in figures 5a and 5b does not



Figure 5a. Box plots of radon concentrations as determined by emanation versus liquid scintillation analysis.



Emanation



Liquid scintillation

Figure 5b. Relative frequency histograms of radon concentrations as determined by emanation versus liquid scintillation analysis

indicate great differences in radon concentrations between the two datasets. However, the histograms indicate that the data for both analytical techniques are not normally distributed. In addition, the box plots show that there are outlying data points present. The box in the figure represents the interquartile range (IQR) or the middle 50 percent of the data. The line across the box is the median value (50th guartile), the ends of the box represent the 25th and 75th quartiles. The length of each stem represents up to 1.5 times the IQR, depending on where values fall in this area. Shorter stems result when data points are lacking in the stem region. Values extending beyond the stems are called outlying data points. Outliers may be present regardless of the length of the stem. The nonnormal distribution of the data violates an assumption of the classical statistical test employed in this section to compare the two analysis techniques. This indicates that a transformation of the data or other robust method of statistical analysis will be needed. The hypothesis testing section will describe this in greater detail.

Figure 6 shows a scatter plot of radon concentrations measured in the groundwater of the fifty two sites analyzed by both emanation and liquid scintillation counting methods. If the two analysis procedures produce similar results for the same sites sampled, then the slope of the best fit line through the data in the scatter plot should be equal to one. Simple linear regression of the liquid scintillation data



Radon-222 concentration by emanation (pCi/L)



(dependent variable) on the emanation data (independent variable) yields a slope of 0.94 indicating that on the average the emanation results are slightly higher. A Pearson correlation matrix for the two variables generated a correlation coefficient of 0.98, which is highly significant.

The other primary objective of this study was to determine whether there were significant differences in radon concentrations in groundwater sampled from different geological formations. Figures 7a and 7b show the radon data grouped in five geological classifications for emanation and liquid scintillation counting analysis respectively. Examination of the grouped data shows nonnormal distributions (note off center locations of medians), outlying data and inequality of variance between groups (note different length boxes). Again, because the data do not satisfy test assumptions well, robust methods will be used in the next section to test the influence of geology on radon concentration.

Hypothesis testing

Hypothesis tests are used to answer experimental questions on a statistical or probabilistic basis, and to make inferences about a population based on a sample from the population. Each question is expressed in the form of two hypotheses concerning a population parameter selected to represent the population. The hypothesis tests establish a null hypothesis and an alternative hypothesis, and then



Figure 7a. Box plots of radon concentrations, grouped by rock type, as determined by emanation analysis. Rock types: CP=coastal plain, GN=gneiss & schist, GR=granite, MA=mafic, MV=metavolcanic.

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Rock types

Figure 7b. Box plots of radon concentrations, grouped by rock type, as determined by liquid scintillation analysis. Rock types: CP=coastal plain, GN=gneiss & schist, GR=granite, MA=mafic, MV=metavolcanic.

produce a test statistic calculated from parameters of the sample. The test statistic is compared to a tabulated critical test statistic of a specified confidence level, which assumes the null hypothesis. The decision to accept or reject the null hypothesis is made based on whether the sample statistic exceeds the critical value. Before a test can be performed, certain prior conditions must be satisfied as nearly as possible. Checking to see if these conditions were met was the purpose of the exploratory analysis, which revealed that certain assumptions were not well satisfied.

The question which asks whether liquid scintillation analysis produced results comparable to standard emanation analysis is tested by looking at the differences between the two measurements for each site sampled. A one sample paired T-test is used because the samples were collected at each site in pairs (Koopmans 1987). The null hypothesis states that the mean difference (in the population) equals zero, meaning the two techniques yield the same results. The alternative states that the mean difference does not equal The test statistic is calculated using sample zero. estimators of the population parameters. Exploratory analysis of the fifty two pairs of emanation and liquid scintillation data in figures 5a and 5b indicated the need for a modified hypothesis testing method. Figure 8 shows a box plot of the differences in radon concentrations measured by the two analytical techniques. The outlying data points at each end can have disproportionate influence on the mean



Figure 8. Box plot of the differences in radon concentrations measured by emanation versus liquid scintillation analysis. Differences were calculated by subtracting the liquid scintillation result from the emanation result. of the differences and therefore can reduce the power of the T-test. Trimming outliers from the dataset before performing the test restores much of the power (Koopmans 1987). The outliers are trimmed so that the data better fit the conditions of the hypothesis test used. The term outlier is being used to describe data points which deviate from the mean to a much greater degree than most of the data. It is not implied that outliers are data which are suspected of being the result of improper experimental analyses. The T-statistic calculated from the trimmed data is shown below.

$$T = \frac{\overline{x}_{T}}{\frac{s_{T}}{\sqrt{y_{T}}}}$$
$$T = 2.954$$

where: \overline{X}_{T} = 10% trimmed mean difference = 79.6 pCi/L S_T = trimmed standard deviation = 170.5 pCi/L N_T = trimmed sample size = 40

The critical value at the 95 percent confidence level for this test is 2.042. Since the sample statistic exceeds this, the null hypothesis is rejected with 95 percent confidence. Therefore it is inferred, based on the sample, that there is a significant difference in radon concentrations measured by the two techniques.

Figure 9 shows a box plot of the differences expressed as percents. Despite the result of the paired T-test,



Figure 9. Box plot of percent differences in radon concentrations measured by emanation versus liquid scintillation analysis. Percent difference was calculated as the absolute value of the emanation result minus the liquid scintillation result, divided by the average of the two, all times 100. examination of the percent differences in appendix IV and their distribution in figure 9 shows that only five values are greater than 45 percent difference. Alternatively, forty five of fifty two values are below 35 percent difference and forty values are below 25 percent difference. Although there are statistically significant differences between emanation and liquid scintillation results, the majority of them can be considered relatively small on a practical basis.

With modification of sampling materials and procedures used, these differences could be substantially reduced. Noting the signs of percent difference values in appendix IV, in 75 percent of the cases it is positive, indicating that the emanation result is greater than the liquid scintillation result. The sample collection technique for liquid scintillation analysis could provide insight as to why these results were often lower. During collection a water sample is drawn from a gently flowing, nonaerating pool of water using a syringe. Careful attention must be paid to this procedure. The degree of success achieved in executing this step with minimal radon loss may partially account for the lower liquid scintillation results. By comparison, samples for emanation analysis were collected directly from a faucet, via rubber tubing, into the bubbler. The potential for loss of radon during this collection process is lower. The vials into which liquid scintillation samples were dispensed may have contributed to lower results

as well. Glass vials and plastic caps with paperbacked foil liners were used in this study. Vial caps with better sealing capabilities would reduce the likelihood of radon loss due to leakage. It is also important to keep in mind that the emanation result for each site is based on only one sample while each liquid scintillation result is the average of two measurements. Although it would require a significant amount of extra time for analysis, it would be best to obtain an average emanation result for each site. By taking the following measures, substantial reductions in the differences in radon concentrations measured may be possible. Strict attention must continue to be given to liquid scintillation sampling technique. To further reduce the potential for radon loss after sampling, improved collection vials should be used. Finally, an average result for both analytical techniques should be obtained for each site.

The other objective of the study, which asks whether site geology has an influence on radon concentration, is tested by looking at the variation in concentrations between the five geological groups sampled. This is accomplished using one way analysis of variance (Koopmans 1987). The null hypothesis states that there is no difference in mean radon concentration between any of the five groups. The alternative is that there is a difference between at least two of the groups. Exploratory analysis of the data in figures 7a and 7b indicated the need for a modification of

the data prior to testing. In this case, the data are transformed by taking their natural logarithms. Figures 10a and 10b show the effect of the transformation on the data. The data fit the test assumptions better, however outlying data points persist in a number of the groups. Applying the trimming technique to each group of the emanation data improves the power of the test further. The analysis of variance tables in table 7 show significant differences between groups for both the emanation and liquid scintillation counting data (the null hypothesis is rejected in both cases). The liquid scintillation counting data were not trimmed following the transformation because of the lack of enough data in each group (see figure 7b for group sample sizes). The small sample sizes in some of the groups as well as the variation in sample size between groups of the liquid scintillation data has undoubtedly affected the power of the analysis of variance test.

The analysis of variance indicates that for both emanation and liquid scintillation analysis at least two groups have different mean radon concentrations. In order to tell which groups are different, each mean is compared to the other four individually by a least significant difference method (Koopmans 1987). Results of these individual comparisons are depicted in figure 11. The rock groups are arranged in order of increasing mean radon concentration. Groups that are inside sets of brackets were not significantly different. All other pairs of groups were



Figure 10a. Box plots of natural logarithms of radon concentrations, grouped by rock type, as determined by emanation analysis. Rock types: CP=coastal plain, GN=gneiss & schist, GR=granite, MA=mafic, MV=metavolcanic.



Natural log of radon concentration by liquid scintillation

	sum of squares	degrees freedom	mean square	F-statistic	p-value
between groups	64.1	4	16		
within groups	70.2	71	0.99	16.211 (reject nul)	0.001 1 hypothesis)

Dependent variable: In emanation result

Dependent variable: In liquid scintillation result

	sum of squares	degrees freedom	mean square	F-statistic	p-value
between groups	20.4	4	5.1		
within groups	44.5	47	0.95	5.385 (reject null	0.001 hypothesis)

Table 7. Analysis of variance tables for emanation and liquid scintillation analysis.





Liquid scintillation

CP	MV	МА	GR	GN
Low radon-222			Hi	gh radon-222
CP	MV	These two groups we	ere <u>not</u> significa	antly different.
MV	ма	These two groups we	ere <u>not</u> significa	antly different.
_ ма	GR	GN These three different.	e groups were <u>no</u> t	t significantly

Figure 11. Results of least significant difference groupings for emanation and liquid scintillation analysis. Rock groups are arranged by increasing mean radon concentration. Sets of brackets are intended only to depict which rock groups were <u>not</u> significantly different. Therefore, other combinations or pairs of groups were significantly different.

significantly different. Liquid scintillation precision

Two samples were collected at each site for liquid scintillation analysis. This provides the opportunity for testing the precision of liquid scintillation analysis by applying a paired difference T-test to the data. The results of the two measurements for each site are listed in table 8. A one-sample paired T-test was performed on the differences between the two measurements obtained for each site. The null hypothesis tested states that the mean difference equals zero. The difference data are shown in figure 12. The figure shows that the data contain outliers, so it was first trimmed before applying the test. The T-statistic calculated from the trimmed data is shown below.



where: \overline{X}_{T} = 5% trimmed mean difference = 8.7 pCi/L S_T = trimmed standard deviation = 64.7 pCi/L N_T = 46

The critical T-value at the 95 percent confidence level is 2.021. Since the sample statistic is less than 2.021, the null hypothesis is accepted with 95 percent confidence. Therefore it is inferred, based on the sample, that there is no significant difference in radon concentrations measured in paired liquid scintillation samples.

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L.S. sample 1	L.S. sample 2	Pent diff.
342 + 17 7	370 + 17 7	77
675 + 10 5	651 + 10 4	3.6
202 + 16 5	331 + 16 7	12.5
292 1 10.5	152 ± 17.2	12.5
254 ± 18.2	152 ± 17.3	50.0
325 + 18.2	539 ± 19.7	49.4
691 ± 18.5	709 ± 18.8	2.0
2002 ± 27.7	1816 ± 26.0	9.8
1593 ± 25.1	1498 ± 24.5	6.1
1958 + 27.5	2327 ± 30.8	17.2
1113 + 23.0	1185 ± 23.2	0.3
129/ ± 24.1	1241 ± 23.8	4.5
2842 ± 35.9	2832 ± 35.8	0.4
1698 ± 26.2	1659 ± 26.0	2.3
891 ± 22.1	926 ± 22.4	3.8
1362 ± 28.8	1397 ± 29.0	2.6
288 ± 22.7	340 ± 23.0	16.7
1215 ± 21.8	1245 ± 22.0	2.5
7949 ± 85.0	7644 ± 82.0	3.9
2241 ± 29.2	2121 ± 28.1	5.5
2065 ± 27.6	2086 ± 27.8	1.0
1264 ± 22.6	1170 ± 22.1	7.7
484 ± 18.3	516 ± 18.6	6.4
3432 + 40.8	3330 ± 39.8	3.0
284 + 17.2	311 + 17.4	9.0
1238 ± 22.9	1276 ± 23.0	3.1
1114 ± 22.3	1083 ± 22.1	2.8
1018 + 21.9	1024 ± 21.9	0.6
1063 ± 22.2	1103 ± 22.5	3.7
359 ± 19.4	323 + 19.2	10.4
175 ± 18.3	186 + 18.4	5.7
4949 + 56.4	4834 + 55.3	2.4
3193 ± 39.4	3119 + 38.7	2.3
1532 + 22.4	1460 + 22.1	4.8
452 + 16.7	497 + 17.1	9.6
9879 +103.7	9942 +104.4	0.6
2013 + 28.1	1975 + 27.8	1.9
1553 + 25.7	1439 + 25.0	7.7
238 + 18.2	234 + 18.2	1.7
2693 + 34.3	2762 + 35.1	2.5
4298 + 49.8	4524 + 52.0	5.1
2648 + 34 1	2620 + 33 7	1 1
285 + 15 4	267 + 15 3	6.7
10 + 23 6	52 + 23 6	5.5
62 + 22 9	94 + 24 0	40.6
942 + 29.0	94 <u>+</u> 24.0	1.8
104 + 15 1	01 ± 14 0	13 1
2165 + 40 0	2072 + 40 4	3.0
3105 T 49.9	1040 + 44 0	3.0
1907 + 44.4	1940 1 44.2	2.0
1125 ± 20.3	1090 + 20.0	3.2
417 ± 19.2	484 ± 19.7	14.9
722 ± 20.5	690 ± 20.3	4.6
1419 ± 24.8	1486 ± 25.2	4.6

Table 8. Radon concentrations determined by liquid scintillation analyses of two sample vials collected at 52 sites. Pcnt diff = percent difference.

Figure 13 shows a box plot of the differences expressed as percents. It should be noted that only three of the values are above 40 percent difference, while forty nine of the fifty two values are below 20 percent difference and forty three values are below 10 percent difference. These results show good reproducibility in the paired liquid scintillation measurements. This is valuable information to have since it lends credibility to the measurement technique. The relatively few cases of larger percent differences may be partially attributable to leakage of radon from one vial of some of the pairs. Looking at table 8, it seems that the greatest percent differences are associated with lower radon concentrations, indicating that the difference expressed as a percent is large because the concentration is low. However, this is not strictly the case. A number of sample pairs at low concentrations have low percent differences between them. Figure 14 shows a plot of the differences between radon concentrations measured in dual LSC samples versus the mean of dual LSC samples.

In general, the results of hypothesis tests should be taken with some reservation. Factors which can affect test validity and power should be considered. This is particularly true when considering the effect of a variable, such as geology, on measured radon concentrations. Since inferences are made about the population based on small samples, anything which could influence how representative



Figure 12. Box plot of the differences between radon concentrations measured in dual liquid scintillation samples.



Figure 13. Box plot of the percent differences between radon concentrations measured in dual liquid scintillation samples.



Figure 14. Plot of differences in radon concentration measured in dual LSC samples versus the mean radon concentration of dual LSC samples.

the sample is can influence the power of the tests. For instance, the way site selection was performed is of great significance from a statistical viewpoint. In addressing the issue of geological influence on radon concentration, within each rock group sites should be selected in a completely random manner with equal probability of selection for each one. A sampling problem exists in the liquid scintillation data because of the fact that samples were not collected at all of the sites. This resulted in a nonrandom sample and unbalanced sample sizes between rock groups, both of which can effect the power of the analysis of variance During sample collection and analysis, systematic test. error should be minimized by emphasizing consistency in procedure. In this study, only one sample was collected from each site for emanation analysis. Collecting two or more to obtain an average for each site would improve statistical power. Sampling was carried out over the course of about five months, including a summer during which the southeast experienced a severe drought. The potential effect of the water shortages or the time span itself on measurements is something that can only be speculated about, but which may have had a significant influence on results. Lower limit of detection

A measure of the detection capability of analytical equipment and procedures is the lower limit of detection (LLD). The LLD of a sample counting system is primarily dependent on the standard deviation of the background of the

system (Currie 1968). The relationship for calculating LLD is shown below.

LLD = 1.695 x 2 x $\sqrt{2}$ x s_{bkg} x K where: 1.695 = critical Z-value at 95% confidence s_{bkg} = standard deviation of background (cpm) s_{bkg} = 0.19 cpm for emanation analysis s_{bkg} = 0.95 cpm for LSC analysis K = factors converting cpm to pCi/L

The LLD for emanation analysis is 10.6 pCi/L, and for LSC analysis it is 40.6 pCi/L.

Quality control

Quality control measurements were performed on the electronic counting system for emanation analysis on a daily basis when the system was in use. A standard activity reference scintillation cell containing a long lived alpha emitter (not radium-226) was counted in the same way that sample scintillation cells were counted. This was done for the purpose of monitoring the variation in counts obtained with the electronic counting system. Ten one minute counts were taken successively and averaged to arrive at a daily value. These daily values were used in a chi-square goodness of fit test. If the system is functioning properly then the relative frequency distribution of the daily values should fit a poisson distribution (Knoll 1979). The chi-square test statistic calculation is shown below.

 $x^2 = (N-1)s^2 / \overline{x}$ $x^2 = 258$ where: N = number of data points = 56 s = standard deviation = 334.3 counts \overline{x} = mean of the data = 23732.6 counts

This chi-square value exceeds the critical value, which indicates that there was more fluctuation in the counting system than can be accounted for by random statistical variation. The plot in figure 15 shows the daily values plotted versus time. There is a definite pattern in the data about the mean value drawn at 23,732 cpm. This supports the x² test result because a uniform distribution (no pattern) of the data about the mean line would be expected if the fluctuation in the counting system could be accounted for by statistical variation alone. From these observations one could conclude that the performance of the counting system is suspect. However, it should be noted that the percent fluctuation of the daily quality control values about the mean is low (average = 1.2 percent). Although figure 15 shows evidence of inconsistent performance of the counting system, the percent fluctuation from the mean was never greater than 2.9 percent.



Time (days)

Figure 15. Plot of daily quality control values for scintillation cell electronic counting system. Daily values are in cpm. Mean value equals 23732 cpm. Time axis is not to scale.

CONCLUSIONS

The performance of the liquid scintillation technique in comparison to the emanation technique was not as expected. This is reflected in the paired difference T-test, which found a significant difference between measurements obtained using the two techniques. In seventy five percent of the cases the liquid scintillation measurement was lower. The reason for this may involve the liquid scintillation sampling technique itself, as well as the sample vials used. During collection a sample is taken from an open pool of water, therefore particular attention must be given to this step in order to minimize radon loss. The foil vial cap seals used during collection may be a significant source of radon leakage. In future work of this type these vial caps should be replaced with ones that are capable of forming better seals. Caps with cone shaped plastic liners are recommended. More than one emanation sample should be collected in order to obtain an average result to compare with an average liquid scintillation Although there were statistically significant result. differences between radon concentrations measured by the two analytical techniques, the percent difference data in appendix IV show that most of these differences are of a tolerable magnitude considering the early stage of implementation of the liquid scintillation analysis technique. Forty of fifty two values are below 25 percent difference and only five values are above 45 percent

difference. This was a first attempt at using the LSC method in the Radiological Hygiene Laboratory and the results obtained with this method were not as expected. Quality assurance liquid scintillation analyses of samples of known radon concentration would be useful for cross checking the accuracy of the system. Future efforts aimed at refining this newer method should prove more successful with the aid of suggestions made in this report. More work with the liquid scintillation method is warranted considering the advantages of this technique over emanation analysis. The liquid scintillation analysis procedure is automated and consequently is much less time and labor intensive than emanation analysis.

The results of liquid scintillation analyses on dual LSC samples collected showed good reproducibility. The paired difference T-test on the dual LSC measurements indicates that the mean difference between dual samples is equal to zero. Forty three of fifty two of the differences are below 10 percent difference and forty nine are below 20 percent difference. The liquid scintillation calibration efficiency (10.86 cpm/pCi) was in good agreement with the expected value of 10 \pm 1 cpm/pCi (Larry Kanipe, personal communication). One hundred percent efficiency would correspond to 11 cpm/pCi (Prichard 1977).

The analysis of variance performed on the grouped data from both analytical methods indicates that there is a difference in mean radon concentration in groundwater

sampled from the five geological groups. The box plots in Figures 7a and 7b show this guite well. This strengthens the hypothesis that geology can be used as a predictive variable for radon concentration in groundwater. Samples from the gneiss/schist and granite rock groups contained the highest levels of radon, the mafic and metavolcanic groups showed intermediate levels and samples from the coastal plain region contained the lowest radon concentrations. The least significant difference groupings in figure 11 show that for both analytical techniques, the grouping test categorized together the mafic and metavolcanic as well as the granite and gneiss/schist groups. But in the case of the liquid scintillation data, other groups were categorized together as well (the mafic, granite and gneiss/schist groups). This indicates that some significant differences were not detected by LSC analysis, while they were detected by the emanation analysis method. This discrepancy in the least significant difference groupings can be partially attributed to the unbalanced sample sizes representing the five rock groups of the liquid scintillation data. A more complete set of liquid scintillation measurements would give the analysis of variance and least significant difference tests much more power.

APPENDIX I

Settings for scintillation cell counting electronics

Canberra model 1400 NIM bin and high voltage supply were powered through an SGL Waber model 24 multiple outlet strip.

The preamplifier, amplifier, single channel analyzer and counter-timer were powered through the NIM bin.

High voltage = 1000 volts

Coarse gain = 50 Fine gain = 1 Input mode dial = differential negative Integrate dial = out Differentiate dial = 2 microseconds Unipolar output range dial = 10 volts negative BLR dial = out Input to amp at normal input jack Output from amp at unipolar output jack

Output from SCA to positive scaler input jack Upper level discriminator (window) = open Lower level discriminator = 0.5 Set to dual disc

Adjust timer settings as needed

APPENDIX II

Procedure for liquid scintillation analysis

Sampling equipment

1. A 20 ml syringe with 18 gauge needle (large bore).

2. Sampling funnel about 4 inches in diameter.

3. Threaded faucet adaptor.

4. Several feet of clear plastic tubing to connect adaptor and funnel.

5. 20 ml liquid scintillation counting vials with lined screw on caps.

 Mineral oil scintillation fluid (High Efficiency, order # PSS-007H, New England Nuclear, Pilot Chemicals Division, 575 Albany St., Boston, MA 02118).

Sampling procedure

1. Prepump well several minutes.

 Locate a faucet for sampling as close to the wellhead as possible, make certain it does not draw water from a storage device.

3. Connect threaded faucet adaptor.

4. Attach clear plastic tubing to adaptor.

5. Fit free end of tubing to funnel.

Turn on faucet and allow water to flow until all air is purged from the collection hose.

7. Holding funnel upright, reduce water flow rate allowing an overflowing pool to form in the funnel without aeration or significant turbulence.

8. Insert tip of hypodermic needle about 1 to 2 inches below surface of water, draw several milliliters into the syringe and discard water (repeat a few more times).

9. Again immerse the needle and very slowly draw slightly more than 10 ml of water into the syringe while attempting to minimize turbulence in the sample.
10. With syringe pointing upward carefully eject air and excess water to obtain a final sample volume of 10 mls.

11. Submerge needle to the bottom of a scintillation counting vial already containing 10 ml of cocktail. Slowly eject water at the bottom of the vial while attempting to minimize turbulence in the sample.

 Tightly cap vial, label appropriately and note time of collection.

13. Immediately collect another sample repeating steps 6 through 12.

NOTE: It is important to have minimal airspace inside the capped vial after sample collection. Radon escaping into such an airspace would not be counted during processing. Adjust volumes used accordingly.

Counting equipment

- 1. Standard activity vials.
- Background vials.

 Packard Tri-Carb 300 liquid scintillation counter. program settings:

Terminators: minutes=50, % deviation=2 Radionuclide=manual Windows: A: LL=0 KeV UL=2000 KeV B: LL=5 KeV UL=1850 KeV C: LL=0 KeV UL=5 KeV QIP=yes AEC=no SCR=A/B # vials/std=1, #vials/sample=1, # counts/vial=1 BKG=manual: A=0, B=0, C=0 % of standard=no low cpm reject: A=0, B=0, C=0 Divide factor K=1 Data mode=cpm

NOTE: The actual counting window is B (5 to 1850 KeV). Use counts from this window as gross cpm to calculate radon concentration.

Counting procedure

1. Preprogram counter with program settings above.

Prepare at least two background vials using distilled water in place of sample water. 3. Set appropriate number program marker to count a set of samples twice and insert the marker at the front of a sample holding tray.

4. Shake all vials to be counted (background, standard and sample vials) for about 15 seconds to thoroughly mix the aqueous and organic phases. Check for leaks around caps. Liquid phases will reseparate quickly.

5. Load background vials in front, standard activity vials next and pairs of sample vials last in sample holding tray. Use additional trays directly behind first tray if there are more than 10 vials.

 Load trays in counter, initiate counting by simultaneously depressing the "forward" and "enable" spots on the key pad.

7. As trays are advanced toward the pickup point observe to make certain that the first vial is picked up. Also make certain you understand the order in which vials will be counted (write it down) and the time and day the first vial began counting.

8. The entire set of vials should count through twice with no periods of interruption between series of The second set of counts are used in s. This allows time for radon daughter in counts. calculations. equilibration inside the vials. Alternatively, set Start the vials aside for four hours after shaking. counter at a time when it is unlikely someone will interrupt it because you need to be able to infer the midpoint of counting time for each vial based on when the first vial started and the elapsed time the counter has been counting under the program as listed on the printout.

APPENDIX III

Procedure for aqueous radium-226 standard dilutions

All work was done in a fume hood on absorbent paper while wearing rubber gloves. Afterward, all materials were monitored with a thin end window GM tube. No contamination was detected.

Three ampules of aqueous radium-226 (4.45 nanoCi/gram) were received from the E.P.A. in Las Vegas, Nevada. Each ampule came with a certificate of calibration listing total activity, volume and mass. The total volume of each was 5.35 ml (5.35 gram), and the total activity in each ampule was 23.8 nCi.

1. Four ml of 1.6M HNO₃ (nitric Acid) was diluted to 60 ml with distilled water. This acid solution was used to thoroughly rinse the insides of a 100 ml volumetric flask and a 100 microliter Eppendorf pipette tip.

2. Most of the acid was then poured into the volumetric flask.

3. The sealed ampule of radium was then broken open and the contents poured into the volumetric flask.

 Some of the remaining uncontaminated acid was pipetted into the ampule using the Eppendorf pipette.

This acid rinse was poured into the flask, followed by all the remaining unused acid solution.

 Enough distilled water was added to bring the total volume of the solution in the flask up to the graduated mark.

The flask contained 23.8 nCi radium-226 in 100 ml distilled water and 0.064M HNO3 . This corresponds to a concentration of 238 pCi/ml. The acid was necessary to prevent excessive plating of radium on the insides of the flask. The ampules received from the E.P.A. contained 0.05M HNO3 . The final dilutions were made as follows.

7. A fresh 0.064M HNO 3 solution was made from 1.6M stock.

8. The acid was used to rinse two thoroughly cleaned emanation bubblers, five liquid scintillation counting vials and a 1 ml glass pipette. The acid rinse was discarded. 9. The first bubbler (labelled #2) was then prefilled with 14 ml distilled water and the other (labelled #3) with 13 ml distilled water.

10. Each vial received 10 ml scintillation fluid (mineral oil based, from New England Nuclear) and then distilled water, decreasing in 1 ml increments, such that 9 ml went into the first vial and 5 ml went into the fifth vial.

11. The solution in the volumetric flask was agitated and using the 1 ml glass pipette, 1 ml aliquots were dispensed to bring the total volumes of the bubblers and vials to 15 and 20 ml respectively. The volumes and activities in each standard are given below.

Bubbler #2		Bubbler #3		
14	ml water	13	ml	water
1	ml radium	2	ml	radium
238	pCi	476	pC:	i

Vial #1 Vial #2 Vial #3 Vial #4 Vial#5 10 ml scint 10 ml scint 10 ml scint 10 ml scint 10ml 8 ml water 7 ml water 9 ml water 6 ml water 5ml 1 ml Ra 2 ml Ra 3 ml Ra 4 ml Ra 5ml 238 pCi 476 pCi 714 pCi 952 pCi 1190 pCi

12. The tops of the bubblers were put in place (stopcocks closed) and silicone sealant was applied to the exterior of the joint.

13. Silicone was applied to the threads and liner inside each vial cap before capping the vials.

14. All standards were labelled with nuclide and activity.

APPENDIX IV

Rock type	Rn-222: emanation (pCi/L)	Rn-222: liquid scint (pCi/L)	Pent diff
CP	1693 ± 25.8		
CP	911 ± 37.5	850 ± 29.0	+7.0
CP	656 ±16.9		
CP	598 ± 17.2		
CP	372 ± 13.3		
CP	216 ± 10.1		
CP	202 ± 10.8		
CP	152 ± 8.7		1.1
CP	126 ± 7.6		1.1.1.2.1
CP	107 ± 6.9		
CP	106 ± 13.8	78 ± 23.9	+30.4
CP	106 ± 6.6		
CP	104 ± 7.1		
CP	92 + 7.1		
CP	89 ± 13.6	50 + 23 6	+56 1
CP	89 + 6.7	50 -2510	100.1
CP	87 ± 6.1		
CP	85 + 6.6		
CP	62 + 5.5	•	1.00
CP	54 + 5.9		
Cr	54 _ 5.9		
GN	3424 ± 78.0	3119 ± 49.7	+9.3
GN	3376 ± 74.7	3156 ± 39.0	+6.7
GN	3032 ± 78.8	2728 ± 34.7	+10.6
GN	2906 ± 67.7	1453 ± 25.0	+66.7
GN	2729 ± 32.5		
GN	2403 - 72.8	2181 ± 28.6	+9.7
GN	2221 ± 61.5	2142 ± 29.1	+3.6
GN	2172 ± 62.4	1909 ± 26.8	+12.9
GN	2133 ± 64.3	2634 ± 33.9	-21.0
GN	2110 ± 65.4	2076 ± 27.7	+1.6
GN	2090 ± 61.4	1994 ± 27.9	+4.7
GN	1793 ± 60.6	1545 ± 24.8	+14.8
GN	1636 ± 52.1	1496 ± 25.3	+9.0
GN	1570 ± 55.5	1269 ± 23.9	+21.2
GN	1515 ± 57.1	1679 ± 26.1	-10.2
GN	1273 + 45.5	1149 ± 23.1	+10.3
GN	1108 ± 47.4	909 + 22.3	+19.7
GN	594 ± 31.2	700 + 18.7	-16.4
GN	229 + 21.5	236 ± 18.2	-3.3
GN	3252 + 77.6	4411 + 50.9	-30.2
GN	310 + 22 0	451 + 10 4	-36.0
CN	18585 + 202 0	451 - 19.4	-30.9
CN	10505 - 203.0	1050 + 44 0	
GN	1902 - 57.1	1938 - 44.3	+0.2
GR	25601 ±107.9		
GR	20237 ± 91.9	•	
GR	7836 ± 120.7	7797 土 83.5	+0.5



5490 ± 50.7 GR 5337 ± 50.2 GR 4777 ± 45.0 GR GR 3296 ± 93.0 2837 ± 35.8 +15.0 1609 ± 25.6 GR 1306 ± 46.8 1230 ± 21.9 +6.0 GR 1252 ± 44.8 1108 ± 26.2 GR +12.3 1034 ± 20.3 GR 986 ± 21.3 GR 663 ± 19.5 904 ± 39.1 +30.6 GR 503 ± 29.4 GR 312 ± 16.6 335 ± 23.4 +7.3 GR GR 134 ± 9.1 99 ± 7.3 GR 20 ± 3.2 GR 9911±104.1 11061 ± 144.4 MA +11.0 3491 ± 75.1 4892 ± 55.9 MA -33.4 2567 ± 31.0 MA 1496 ± 22.3 1416 ± 47.0 MA -5.5 1292 ± 49.4 1099 ± 22.2 MA +16.2 1097 ± 49.3 1083 ± 22.4 +1.3 MA 1071 ± 44.5 1257 ± 22.9 MA -16.0 828 ± 36.8 706 ± 20.4 MA +16.0 599 ± 32.6 1217 ± 22.4 MA -68.1 1021 ± 21.9 MA 574 ± 31.9 -56.0 474 ± 16.9 527 ± 28.8 MA +10.4 405 ± 26.5 341 ± 19.3 MA +17.1 273 ± 21.4 276 ± 15.3 MA -1.1 268 ± 22.2 297 ± 17.3 MA -10.4 MA 181 ± 20.3 180 ± 18.4 +0.1 125 ± 8.5 MA 87 ± 7.8 MA 3901 ± 42.7 MV 3381 ± 40.3 +0.7 MV 3406 ± 76.3 1536 ± 53.5 1380 ± 28.9 MV +10.7 1396 ± 25.6 MV 1089 ± 20.2 MV 1059 ± 22.6 MV 1024 ± 21.2 MV 847 ± 20.5 MV 677 ± 35.8 432 ± 19.0 MV +44.2 655 ± 35.5 MV 203 ± 17.7 611 ± 34.7 MV +100.2 596 ± 16.1 MV +10.3 500 ± 18.4 554 ± 30.5 MV 554 ± 15.0 MV . 544 ± 14.9 MV 356 ± 17.7 501 ± 15.5 MV +33.9 482 ± 13.4 MV 314 ± 22.8 356 ± 23.3 MV +12.5 289 ± 10.5 MV +17.0 98 ± 15.0 116 ± 14.5 MV

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Appendix IV. Radon concentrations in pCi/L as determined by emanation and liquid scintillation analysis. Rock types: CP = coastal plain, GN = gneiss & schist, GR = granite, MA = mafic, MV = metavolcanic. Percent difference between emanation and liquid scintillation measurements (Pcnt diff) was calculated as the absolute value of the emanation result minus the liquid scintillation result, divided by the average of the two results, all times 100. A plus sign associated with values of percent difference indicates that the emanation result was larger, a minus sign indicates the liquid scintillation result was larger.

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MV MV

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