A PRELIMINARY STUDY OF TEMPORAL VARIABILITY OF ^{222}Rn IN A PRIVATE WELL WATER SUPPLY

Eileen L. Fisher

Dept. of Preventive Medicine and Environmental Health, College of Medicine, Oakdale Campus, University of Iowa
Iowa City, IA

ABSTRACT

Temporal variations of ^{222}Rn in water suggest that one-time sampling of water supplies may not produce a sample that is representative of well water. Information on temporal variability of ^{222}Rn in water supplies near 300 pCi L^{-1} (11 Bq L-1) has important implications for regulation, as well as implications for population risk assessment. Without information on variability of waterborne ^{222}Rn over time, cumulative ^{222}Rn exposure may be either over- or under-estimated. The objective of this study was to investigate the temporal variability of low-level ^{222}Rn in well-water. Daily sampling was conducted March through May 1993, plus two days of hourly sampling in March, on a private well serving three houses and 10 people. ^{222}Rn levels varied daily and hourly by as much as 15% in a well with a mean ^{222}Rn concentration of 884 ± 31 pCi L^{-1} (33 ± 1 Bq L-1). Further research is needed to allow for a better understanding of the amount of temporal variation of low-level ^{222}Rn in public water supplies, factors affecting this variation and appropriate sampling methods for regulatory purposes.

INTRODUCTION

Waterborne ^{222}Rn poses a health threat through both direct ingestion and escape into the air. Epidemiological studies show a relationship between airborne ^{222}Rn and lung cancer (ICRP 1979, NCR 1988, Samet 1989). Animal studies also support a dose-response relationship between lung cancer and ^{222}Rn exposure (ICRP 1979). It is now estimated by the EPA that between 7,000 and 30,000 lung cancer deaths in the United States may be caused by airborne ^{222}Rn exposure (EPA 1992a). The EPA estimates that waterborne ^{222}Rn contributes about 200 deaths per year in the United States with an uncertainty range of 10-2,700 (Crawford-Brown 1992). The estimated health risk from waterborne ^{222}Rn exposure is predominantly based on breathing degassed ^{222}Rn, rather than ingested ^{222}Rn in water.

Based on the contribution of waterborne ^{222}Rn to airborne ^{222}Rn lifetime risks of fatal cancer have been estimated at 10^{-4} and 10^{-3} for ^{222}Rn concentrations of 200 pCi L^{-1} and 2,000 pCi L^{-1} respectively (7 Bq L-1 and Bq L $^{-1}$) (Crawford-Brown 1992). These estimates indicate that ^{222}Rn in water poses an adverse health effect to the public which is significantly greater than other water contaminants (Kelly 1989).

Previous Studies

The average USA groundwater concentration of ^{222}Rn is in the range of 200 pCi L^{-1} to 600 pCi L^{-1} (7 Bq L-1 to 22 Bq L-1) (Longtin 1990). A recent survey by The University Hygienic Laboratory, Iowa City, Iowa, of community water supplies indicates that one quarter will exceed the proposed ^{222}Rn standard in finished water. The range of ^{222}Rn concentrations were from below detection level to 2,546 pCi L^{-1} (94 Bq L-1) (Kelly and Mehrhoff 1993). ^{222}Rn water concentrations in 352 private wells in Iowa have a similar range with an arithmetic mean of approximately 429 pCi L^{-1} (16 Bq L-1) (Field and Kross 1993). Fifty-two percent of the private wells in the Field and Kross study exceeded the proposed standard for public water supplies. These studies indicate many Iowa water supplies will be at or near the proposed limit for ^{222}Rn.

The above studies of ^{222}Rn concentration in water were conducted at one point in time. There have been few studies of the temporal variation of ^{222}Rn in water. The U.S. Geological Survey tested one well in Pennsylvania monthly for 3 years beginning in 1989 and found ^{222}Rn concentrations which ranged from 2,800 to 7,900 pCi L^{-1} (104-292 Bq L-1) (Senior 1989). They report maximum levels in autumn and early winter and minimums in spring. A study conducted by the Connecticut EPA from 1989-1990 (McHone 1990) examined temporal variation in

5 wells with ^{222}Rn average concentrations of 3,500 to 662,000 pCi L^{-1} (130-24,494 Bq L-1). Hourly, daily and weekly sampling schemes were employed. Their results indicate that a single water ^{222}Rn analysis is not necessarily representative of the ^{222}Rn character of the well. ^{222}Rn concentrations in pCi L^{-1} varied by as much as 94 percent for an individual well, with the high well exhibiting more variation. In Maine (1988) wide variations were found in 10 granite bedrock wells over a 3 week period with average ^{222}Rn levels of 2,619 to 758,760 pCi L^{-1} (97-28,074 Bq L-1). The site with the lowest average had a range of 1,420 to 3,090 pCi L^{-1} (53-114 Bq L-1) (Lowry 1988).

Temporal ^{222}Rn data on low-level ^{222}Rn water supplies are scarce. The Iowa Geological Survey conducted a study in 1981-1982 of ground water quality in the Big Spring Basin (Hallberg 1983). A few monthly samples were collected from 24 domestic water wells with average concentrations of 28 pCi L^{-1} - 1500 pCi L^{-1} (1 Bq L-1 - 54 Bq L^{-1}). Twelve samples taken hourly on one site ranged from 230 pCi L^{-1} to 330 pCi L^{-1} (9 Bq L-1 - 12 Bq L-1). Data from these wells indicate variations of 43 to 700 percent.

The temporal variation of ^{222}Rn in Iowa's public water supplies using ground water is not known. The purpose of this preliminary study is to describe ^{222}Rn variations in a multi-household water supply with low-level ^{222}Rn concentrations. Future studies to gain understanding of radon variation in public water supplies will be necessary to develop a sampling plan to accurately characterize ^{222}Rn levels.

MATERIALS AND METHODS

A preliminary study was conducted on a private well serving three houses and 10 people. This well is in the Silurian-Devonian aquifer with a depth of 250-300 feet. Tritium analysis indicates this water is pre-nuclear testing (1963) or at least thirty years old (Bradbury 1991). Investigations into the hourly and daily variation of ^{222}Rn at a point directly beyond the pressure tank were performed as an independent study project. Water samples were collected daily from 3 March 1993 until 31 May 1993 (most in the early morning). In addition hourly samples were collected on one weekday and one weekend day from early in the morning until late evening hours.

Samples were collected using EPA recommended procedures for determination of ^{222}Rn in drinking water. A hose with attached funnel was connected to a faucet and the water allowed to run for 10 minutes. A 10 ml disposable pipet and pipette-pump was used to transfer a 10 ml aliquot to a liquid scintillation vial containing 10 ml of Opti-fluor liquid scintillation fluid. The samples were allowed to equilibrate for three hours, and then counted for 60 minutes in a liquid scintillation counter within 1 day of sample collection.

The calibration of the liquid scintillation counter is performed using a ^{228}Radium Standard, for ^{222}Rn ingrowth, in the same geometry and matrix as the samples. This standard was secured from the EPA and is traceable to the National Bureau of Standards. Each set of analyses was preceded by a standard and two background vials having approximately the same total dissolved solids as the samples. Additional background vials were analyzed following the samples for each analysis with one additional background run for each four samples. Control charts for the standards and the background were maintained to assure the standard did not exceed two standard deviations more often than one in 20 times.

Duplicate samples were taken in all cases. The two sigma percentage error (%2sigma) was calculated using formulas from Vitz 1991. If the relative percent difference (RPD) was less than %2sigma, then the average of the two samples was used, otherwise the maximum. This was done because it is easier to get a low count due to loss of ^{222}Rn than it is to get a high count.

Quality Assurance/Quality Control procedures as outlined in the EPA publication, "Handbook for Analytical Quality Control in Radioanalytical Laboratories" (EPA 1977) were followed during the course of this study. Inter-laboratory cross-checks with The University Hygienic Laboratory, Iowa City, Iowa, on two occasions showed good agreement with RPD of 4.1% and 0.2%.

RESULTS

Daily sampling from 3 March 1993 to 31 May 1993 resulted in 89 samples with duplicates being analyzed for ^{222}Rn. The average RPD for this phase of the study was 3.0% and the average %2sigma was 3.9. The RPD was less than %2sigma for 76% of the daily samples and less than %2sigma plus 10% for 80% of the samples. The Shapiro-Wilk test for normality, as well as the non-parametric Kolmogorov-Smirnov test, indicate daily and hourly ^{222}Rn concentrations are normally distributed (p > 0.2). Daily samples had a mean of 884.3 ± 31.0 pCi L^{-1} (32.7 ± 1.1 Bq L-1) with a range of 804.7 pCi L^{-1} - 940.3 pCi L^{-1} (29.7 Bq L-1 - 34.79 Bq L-1). Figure 1 shows the variation of the daily sampling.
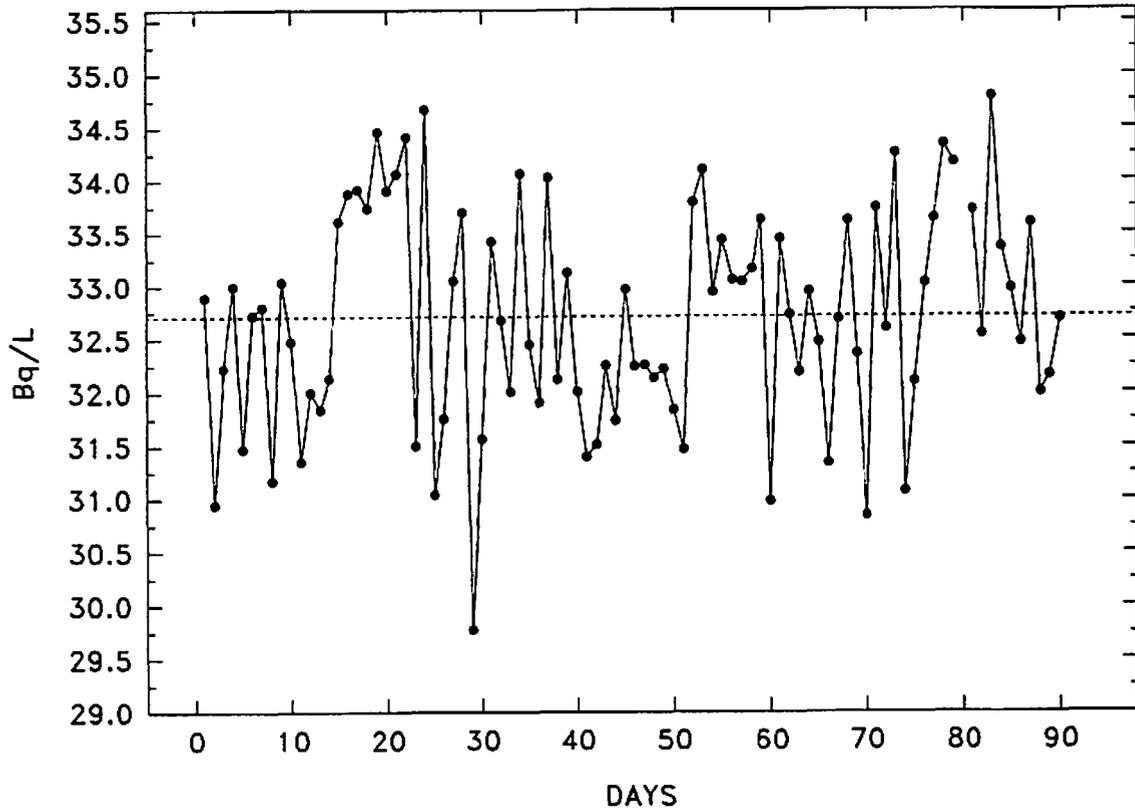

Fig. 1. Daily ^{222}Rn variations in Bq L-1 measured from 3 March 1993 to 31 May 1993. Concentrations measured with a precision of 4% (%2sigma).

Hourly samples on Tuesday, 2 March 1993 had an average RPD of 3.72 and average %2sigma of 4.0. Seventy eight percent were less than %2sigma. Sampling began at 6:30 am and ended at 11:30 pm. The ^{222}Rn concentration varied from 831.3 ± 35.8 pCi L^{-1} to 924.1 ± 35.4 pCi L^{-1} (30.8 ± 1.3 Bq L-1 to 34.2 ± 1.3 Bq L-1). Figure 2 shows the pattern of variation for the first day of hourly sampling. ^{222}Rn levels were lowest in the early hour of the morning, rose to a peak at about 8:30 and then fell and rose again to the highest level occurring around noon.
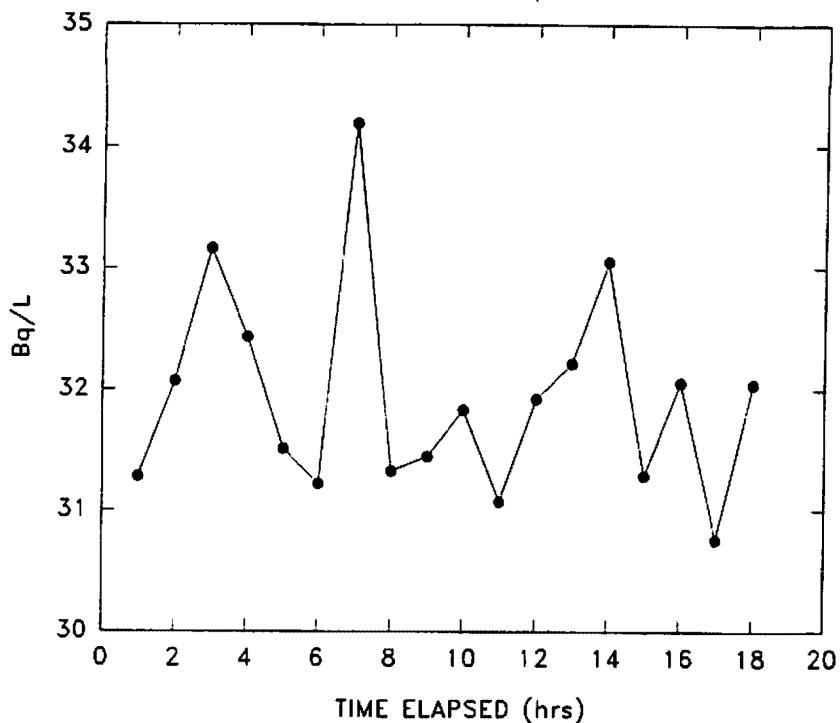
Fig. 2. Hourly [222]Rn variations in Bq L-1 measured on 2 March 1993 (6:30 AM - 11:30 PM).

Hourly sampling on Saturday, 31 May 1993, has a similar range of 847.5 ± 33.9 pCi L^{-1} to 937.4 ± 35.5 pCi L^{-1} (31.4 1.3 Bq L-1 to 34.7 ± 1.3 Bq L-1). Early morning [222]Rn concentrations were again lowest, with a maximum at 11:30 am and another relative maximum at 9:30 pm. The average RPD and %2sigma were 2.7% and 4.1% respectively. Figure 3 is a plot of the [222]Rn concentration verses time.
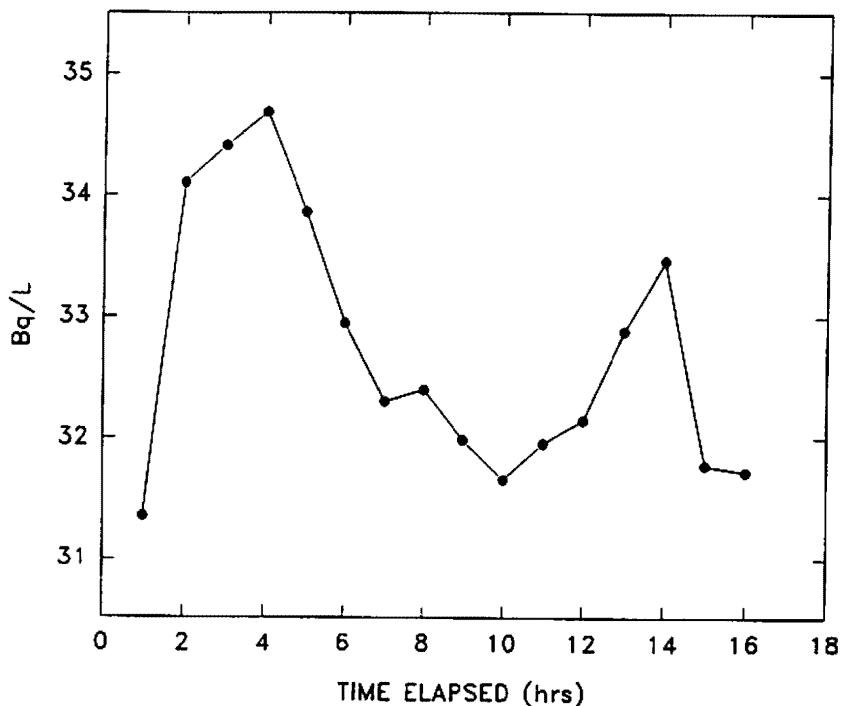

Fig. 3. Hourly [222]Rn variations in Bq L-1 measured on 31 May 1993 (8:30 AM - 11:30 PM).

DISCUSSION

On July 18, 1991, the EPA proposed maximum contaminant levels (MCLs) for radionuclides in public drinking water supplies (EPA 1991). The proposed MCL for waterborne ^{222}Rn is 300 pCi L^{-1} (11 Bq L-1) which corresponds to a risk of 1 x 10^{-4} of death due to lung cancer. Setting a standard for regulation of radionuclides in drinking water has now been delayed until Dec. 31, 1993. There has been much debate over the proposed standard because it is expected that over half of the 60,000 public water supplies in the nation will exceed the MCL for ^{222}Rn. The estimated cost of bringing these water supplies into compliance is $2.5 billion yr^{-1}. More than two thirds of those costs will be borne by the very small water systems that serve 500 or fewer people (Raucher 1992).

The EPA recognizes that ^{222}Rn in water can vary substantially over time. They recommend additional sampling on different days for homeowners when an initial water sample indicates a level of concern (EPA 1992b). This approach does not take into account time of day or seasonal variation. Proposed monitoring in groundwater for public water supplies is quarterly the initial year, then annually unless out of compliance. A system is out of compliance if the average of 4 quarterly samples or one annual sample exceeds the MCL (EPA 570/9-91-700 1991). However, quarterly and annual monitoring may not be the most accurate or economical sampling scheme. Taking an example from the Big Spring Basin study (Hallberg, 1983), the minimum value reported for one site in Feb. 1982 was 70 pCi L^{-1} (3 Bq L-1) with a maximum of 580 pCi L^{-1} (21 Bq L-1) in Mar. 1982. Starting quarterly sampling in January would have resulted in an average of 370 pCi L^{-1} (14 Bq L-1), while starting in February would have yielded an average of 225 pCi L^{-1} (8 Bq L-1). The average of all 11 monthly measurements is 346 pCi L^{-1} (13 Bq L-1), an absolute difference of 24 pCi L^{-1} and 121 pCi L^{-1} (1 Bq L-1 and 5 Bq L-1) from the two quarterly averages respectively.

^{222}Rn concentrations in this multiple household well exhibit variations of as much as 15% over a 3 month period and 11% in hourly sampling. Aging of this water using tritium analysis indicates that the water is at least 30 years old and probably not affected by current climatic conditions (todays' temperature, present year rainfall, wind velocity, barometric pressure) and possibly will show no seasonal or yearly trend. If over time it could be determined that the variation of ^{222}Rn in this aquifer was a stationary process with minimal ^{222}Rn concentrations then monitoring such a system on a frequent basis (yearly) would be unnecessary.

Smaller water supply systems tend to take their water from smaller aquifers which in general have higher ^{222}Rn levels (Milvy 1990). Compliance with drinking water standards places a higher burden on the smaller public water systems which have fewer resources, lack trained staff, and are already economically strained by other factors (The 93' flood). Therefore developing an accurate method for characterizing ^{222}Rn levels in a water supply system or aquifer will be beneficial to small community water supply systems in Iowa. It will ensure that resources devoted to mitigation will be directed at the water supplies that represent the most serious problem.

CONCLUSIONS

Knowledge of temporal variability of ^{222}Rn in water supplies near the EPA proposed MCL has important implications for regulation as well as implications for population risk assessment. Cancer risk assessments are related to cumulative exposure over time and without information on variability of waterborne ^{222}Rn over time, this risk may be either over- or under-estimated. This research suggests that even at low ^{222}Rn concentrations waterborne ^{222}Rn can exhibit substantial variability. More research is needed to determine the variability in public water supplies which will be regulated for ^{222}Rn. If it is found that water ^{222}Rn at these lower levels varies over time then that variation should influence sampling strategies. By avoiding over- or under-estimation of water ^{222}Rn users will be afforded the protection intended by the EPA regulation and unnecessary water treatment for removal of ^{222}Rn will be avoided.

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