A STUDY OF RADIOACTIVITY CONCENTRATIONS IN WARWICK, NY

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ABSTRACT

A project to determine the indoor radon and waterborne radionuclide concentrations in a region of the town was undertaken following the identification of a home containing very high concentrations. Well waters in some area homes were measured four times during the year for gross alpha, gross beta, and dissolved radon levels. Charcoal canisters were provided to participating homeowners to measure indoor radon concentrations. In addition to radionuclide analyses, a few water supplies were analyzed for correlations with other chemical parameters (e.g., trace metals, pH). Though some of the tested wells contained significant radionuclide concentrations, the activity in water supplies did not approach the high radiological concentrations found in the well that triggered interest in this area. As only a few of the homes located on the same geologic formation as the initial home were measured in this project, it is likely that homes with significantly higher waterborne radioisotope concentrations exist in the area.

INTRODUCTION

In January 1998 the State Health Department requested a meeting with representatives from the Orange County Health Department (OCHD) and the Town of Warwick regarding their assistance in collection of well waters from homes in a region in which a well containing high concentrations of dissolved radon, uranium, and radium had been identified by research conducted by a Ph.D. student at SUNY Albany School of Public Health. The region is located in the Hudson Highlands, an extension of Pennsylvania's Reading Prong, that includes parts of Dutchess, Orange, Putnam, Rockland, and Westchester Counties. It is known that these counties contain water supplies with high radon concentrations (1). Generally, the higher radon concentrations in these counties are associated with fault structures and/or metamorphic rocks and Canadian shield material transported during the last glacial period. However, in Warwick it appears that pigmatite intrusions into the carbonate bedrock provide the source material for radionuclides dissolved in the well waters. As high radionuclide concentrations in drinking water supplies pose a significant health risk and nearly 700 homes are built on the carbonate bedrock,

the intent of this study was to determine the concentrations and extent of dissolved radionuclides in wells of this region. Homeowners, selected from a real-property tax listing of single-family homes located near these intrusions, were solicited by the town of Warwick by telephone for voluntary participation. OCHD scheduled collection dates and times with individual homeowners during each collection period. Following each round of sample collections, results were provided to homeowners.

While no regulations exist regarding radionuclide levels in private water wells, the current U.S. Environmental Protection Agency's (EPA) Safe Drinking Water Standards and the NY State Sanitary Code (Part 5), which apply only to public drinking water supplies, specify a maximum contaminant level (MCL) of 15 pCi/L for gross alpha (GA) and 5 pCi/L for combined radium (^{226}Ra and ^{228}Ra). Currently there is no MCL for either uranium or radon in water, although concentrations near 30 and 300 pCi/L, respectively, are under consideration. While ingested radium concentrates in the bone marrow and is a known carcinogen, ingested uranium has been linked to kidney failure. Radon in water poses a health risk from both ingestion and inhalation (following its emanation from the water). Naturally-occurring radon is responsible for over half the radiation dose received by the public from all sources and is estimated to be responsible for 14,000 to 22,000 lung-cancer deaths annually in the U.S. The primary concern from radon in water is that the radon can be transferred to indoor air during use of the water in the home, adding the contribution to the radon levels found in homes from the surrounding soil. It is estimated that a radon concentration of 10,000 pCi/L in water would supply an indoor air level of approximately 1 pCi/L to a home. The EPA's radon in indoor-air action level is 4 pCi/L.

SAMPLE COLLECTION AND PREPARATION

Of the 29 homes participating in the study, 26 completed all four sample collections scheduled and conducted by OCHD. With one exception, each home was served by a single well. Of those reporting, well depths for the homes were from 75 to 670 feet. Many homes used particulate filters with the water supply and 10 homes used a resin-based water softener (salt regenerant). When possible, samples were collected from a spigot prior to any treatment or storage. Collection dates were influenced by results obtained from the student's sampling of other local wells. Samples were collected during three days each round beginning on the following dates:

first round (March 16, 1998)
second round (August 22, 1998)
third round (September 2, 1998)
fourth round (February 22, 1999)

Water samples for GA/GB activity determinations were collected from a cold-water tap using a 2-liter bottle. Samples were mailed promptly after collection and received within two days. Nitric acid preservative was added upon receipt of samples at the laboratory and within 3 days of collection. Generally, a 100-mL aliquot of sample was used in the GA/GB activity determination, although the volumes ranged from 25 to 250 mL, depending on the mass of the suspected particulates. Following EPA Method 900.0, samples were evaporated to <10 mL using a steam bath, transferred to a stainless steel planchet, and taken to dryness at 80 °C in an oven.

Planchets were stored in a dessicator prior to the activity being measured on a low-background gas proportional counter (detection limit ≅ 1 pCi/L for a counting period of 200 minutes).

Water samples for the dissolved radon measurements were collected at the cold-water tap using a method similar to that of Whittaker (2) and detailed elsewhere (3). After removing any aerator and with water flowing, the tap was immersed into the bottom of a funnel. The flow was regulated such that water flowed both from the outlet of the funnel and over its brim. A graduated syringe was used to remove 10 mL of water from the bottom of the funnel and to inject it below the scintillation fluid in a 22-mL glass scintillation vial. Samples were mailed promptly after collection, received within two days, and measured immediately using a liquid scintillation analyzer and then twice again over four days (detection limit ≅ 20 pCi/L for counting periods of 50 minutes).

Samples for dissolved radium (^{226}Ra) were prepared using EPA Method 903.1 which entails evaporating 0.5-1.0 L of water to a volume of approximately 10 mL, placing the concentrate in a sealed glass bubbler, and waiting 20-30 days for ingrowth of radon. The radon was purged from the water into a 125-mL Lucas cell and measured three times over a week-long period using an alpha-scintillation counter (detection limit ≅ 8 pCi/L for counting periods of 100 minutes). Samples for dissolved uranium isotopes were prepared using EPA Method 908.0 which includes electroplating separated uranium isotopes onto stainless steel disks and measuring the activity using a surface-barrier detector (detection limit ≅ 0.05 pCi/L for counting periods of 1000 minutes).

RESULTS AND DISCUSSION

Dissolved Radon Concentration

As shown in Figure 1, all radon concentrations in the wells exceeded 300 pCi/L (an EPA-proposed MCL), with nine wells averaging >3000 pCi/L (a possible alternative MCL being considered by EPA) during the study period. Concentrations ranged from 430 to 21,800 pCi/L in the 29 homes, with all but one well below 10,000 pCi/L. The arithmetic and geometric mean concentrations were 3100 and 2000 pCi/L, respectively. Concentrations in all wells varied within a factor of three over the one-year study period, with no consistent seasonal variability. In addition to natural concentration fluctuations, some variability was inevitably due to different sampling conditions. While an effort was made to collect samples prior to any treatment and from the same taps during the project's duration, records indicate that collection locations and treatment by-passes varied in some homes, possibly accounting for some of the concentration variation. Results of 55 duplicate radon sample collections conducted during the first two rounds showed an average variation of 6% in concentrations. Confidence in representativeness of sample collection resulted in only a single radon sample being collected in the final two rounds of the study.

Gross Alpha (GA) and Gross Beta (GB) Activity

The mass of dissolved and suspended solids varied widely from sample to sample. As the gas proportional detectors are calibrated for sample masses of 100 mg or less, volumes used for GA activity measurements varied from 25 to 250 mL to satisfy that limitation. Sample masses

ranged from 11 to 100 mg, but were generally near 50 mg. A good indicator of dissolved solids (hardness) is the ratio of volume to mass. The ratio was generally below 4 mg/mL, except for one home with ratios near 10. In general, the ratio was highest for the first round of samples collected.

GA activity ranged from 1 to 111 pCi/L in the well waters (Figure 2), with arithmetic and geometric means of 12 and 6 pCi/L, respectively. At these homes, GA activity was comprised of uranium and radium isotopes, as no thorium (^{232}Th) decay products were detected in any samples. During the year, six wells contained GA>15 pCi/L in at least one collection and four wells were consistently above 20 pCi/L. On average, GA activity in all wells varied by a factor of two. There was little correlation (Figure 3) of gross alpha activity and dissolved radon concentrations.

GB activity ranged from 1 to 32 pCi/L in the well waters, with arithmetic and geometric means of 4.7 and 1.3 pCi/L, respectively. At these homes, GB activity is comprised of uranium and its decay products and ^{40}K. During the year none of the wells contained GB>50 pCi/L, specified in the USEPA Safe Drinking Water Standards and in Part 5 of the NY State Sanitary Code as a monitoring compliance limit. On average, GB activity in all wells varied by a factor of two, though concentration variations up to a factor of four occurred.

Isotopic Radium and Uranium Concentrations

Well water from six homes with elevated GA activity (>15 pCi/L) were analyzed in the first round for isotopic constituents of radium and uranium. Water samples from each of three of these homes were analyzed again in the fourth round to examine variability. ^{226}Radium levels in the first round ranged from 0.2 to 16.5 pCi/L, with arithmetic and geometric means of 7.3 and 2.6 pCi/L, respectively. Up to 35% of the GA activity was contributed by ^{226}Ra, although the average value was near 14%. Half of the samples contained ^{226}Ra above the combined (^{226}Ra and ^{228}Ra) MCL of 5 pCi/L specified in the USEPA Safe Drinking Water Standards and Part 5 of the NY State Sanitary Code. There was a positive correlation of ^{226}Ra concentrations with total uranium concentrations. Results from the fourth round were identical to those for the first round.

The range of concentration for each of the natural uranium isotopes, ^{234}U, ^{235}U, and ^{238}U was 7 to 42 pCi/L, 0.5 to 2.2 pCi/L, and 7 to 42 pCi/L, respectively. On average, total U accounted for 91% of the GA activity. Two wells contained uranium at concentrations above 30 pCi/L. Results from the fourth round were identical to those of the first round. Ratios of ^{234}U/^{238}U, generally above unity in natural waters, varied from 1.0 to 1.4 in the wells. ^{235}U concentrations (in pCi/L) were 5 to 7% of those for ^{238}U, approximately as that which occurs naturally.

During the first round, water from one home contained total uranium concentrations near 11 pCi/L, but ^{226}Ra was below the detection limit (0.01 pCi/L). As these isotopes are often correlated, this sample was the subject of additional analyses. Results from trace element analysis (below) indicated no measurable metals (e.g., Ca, Mg, K) were in the water, except for sodium. Reexamination of the drinking water system at the home revealed that a softener was operational and the sample was taken from a tap supplying softened water. From additional studies outside this project it has been observed that water softeners are effective at removal and retention of radium, but have little effect on waterborne uranium concentrations. The results are similar to those we observed in this study. Based on the additional studies it appears that regeneration ('back-flushing') of the softener exchange resin is somewhat ineffective at removing the radium,

and therefore, softeners will gradually accumulate radium activity. For example, a softener processing 500 L (130 gal) of water containing 6 pCi/L of GA activity (10% due to ^{226}Ra) will accumulate 300 pCi/L of radium daily, which, within two weeks, will produce an additional 900 pCi/L of GA activity. In several of the homes in these studies, the GA activity in water following the softener actually exceeded that of the untreated water, indicating the release of long-lived ^{226}Ra decay products from the resin into the water supply. Considering the health implications of this phenomenon and the number of water softeners in use, this aspect warrants additional study under controlled conditions.

Temperature, pH, Alkalinity Measurements

Water temperature was measured at each home during the first three rounds using either a digital or mercury-bulb thermometer immersed in a container of freshly-collected water. While water temperatures averaged 11°C, first-round temperatures ranged from 4°C to 12°C and averaged 4°C less than those of the second and third rounds. Although reported well depths ranged from 75 to 670 feet, there was no correlation between water temperature and well depth.

Water pH values were measured during the second round, with a few repeat measurements conducted during the third and fourth rounds. All water samples had pH values between 7.0 and 8.1, with the exception of two water samples with values below 6.6. For the four homes measured during the third round, pH values increased slightly.

Alkalinity, a measure of water hardness, averaged 250 mg/L and ranged from 85 to 357 mg/L during the second round sampling. These results mimicked those observed in the other rounds.

Trace Elements Concentrations

Water samples from seven homes where elevated GA activity was found were analyzed for trace element concentrations to examine possible correlations. The most abundant elements detected, Ca, Na, and Mg, are representative of the carbonate bedrock in the area. Only Na concentrations exceeded the recommended limits for public water supplies. While no clear pattern exists, GA activity generally was negatively correlated with trace element concentrations.

Airborne Radon Concentrations

Charcoal-canister detectors were provided to each home for an indoor radon measurement. These were deployed (primarily in basements) and returned to the contract laboratory (RTCA) by the homeowners.. Measurements were completed in sixteen homes, with values ranging from 1 to 72 pCi/L. The arithmetic and geometric mean concentrations were 14 and 6 pCi/L, respectively, indicating a log-normal distribution. Ten homes had radon levels above the EPA action guideline concentration of 4 pCi/L, and five homes had levels >10 pCi/L. As shown in Figure 4, there was little correlation of radon in water with indoor radon concentrations.

Remediation Methods

While remediation was not part of this project, homeowner interest resulted in the installation of several point-of-use (POU) water systems in the studied homes. Several homeowners installed commercially-available systems generally costing <$100. However, these are warranteed only for Cu and Pb, and, though effective for some, do not efficiently remove the

dissolved radionuclides. The role of water softeners in removing radioactivity from the water was examined. The most effective treatment systems examined are those utilizing reverse-osmosis technology, though these are costly (roughly $600). These systems remove all measurable uranium, radium, and radon.

CONCLUSIONS

A study of dissolved radioactivity in well waters and indoor air radon concentrations at 29 homes in the town of Warwick, NY, showed that the very high concentrations determined in the well that initiated interest in this area were not widespread in the region. Well waters supplying the homes in this study exceeded 15 pCi/L of GA activity in only six homes, but reaching levels to 111 pCi/L. As only a few (4%) of the homes located on the geologic formation were measured in this project, it is likely that homes with higher waterborne radioisotope concentrations exist in the area. Overall water quality (and quantity) is poor in this region as evidenced by the number of water purification systems installed, the trace metal and radioisotope concentrations, and from interviews with homeowners.

REFERENCES

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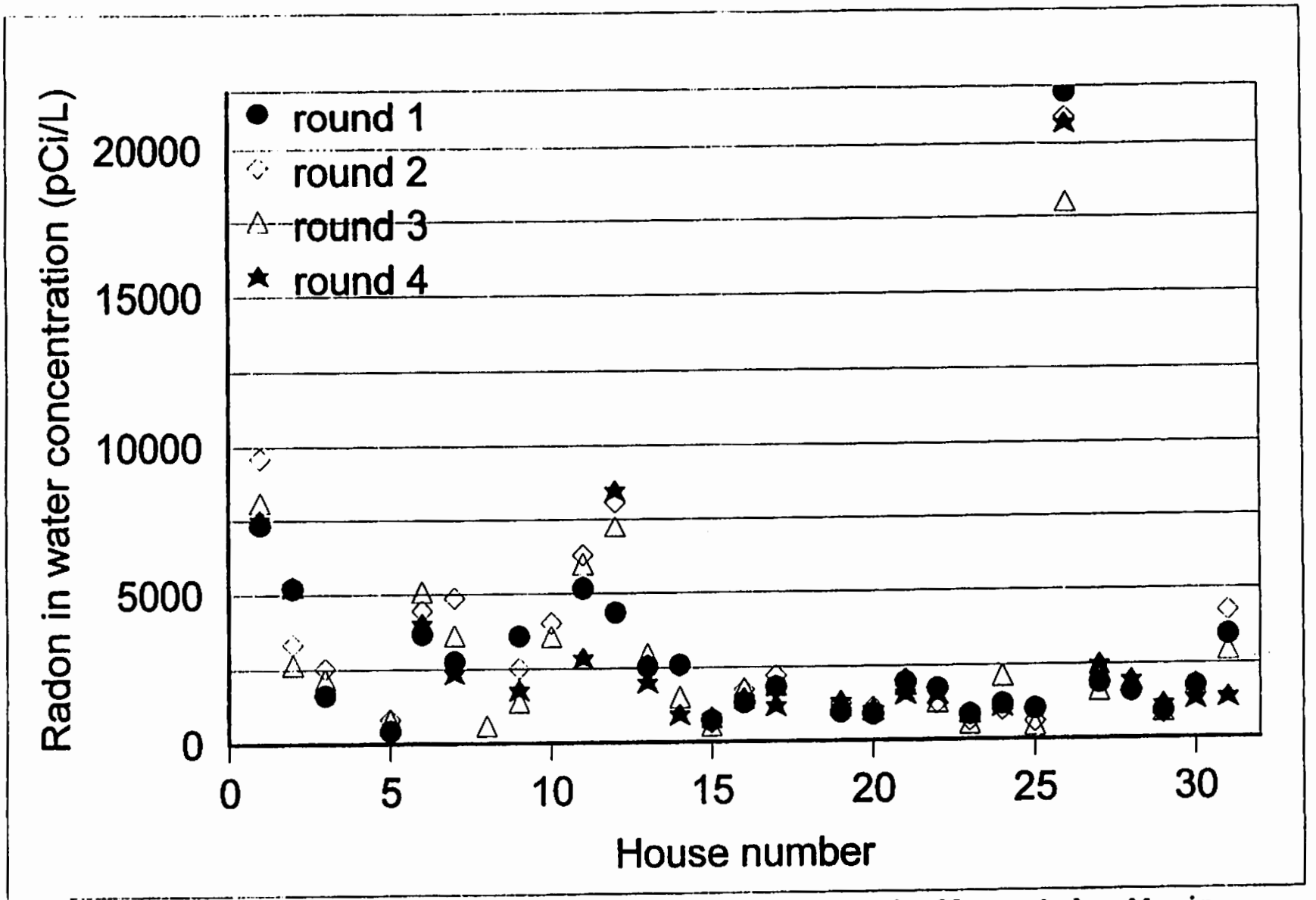

Figure 1. Waterborne-radon concentrations (pCi/L) observed in 29 sampled wells in Warwick during four times of the year.

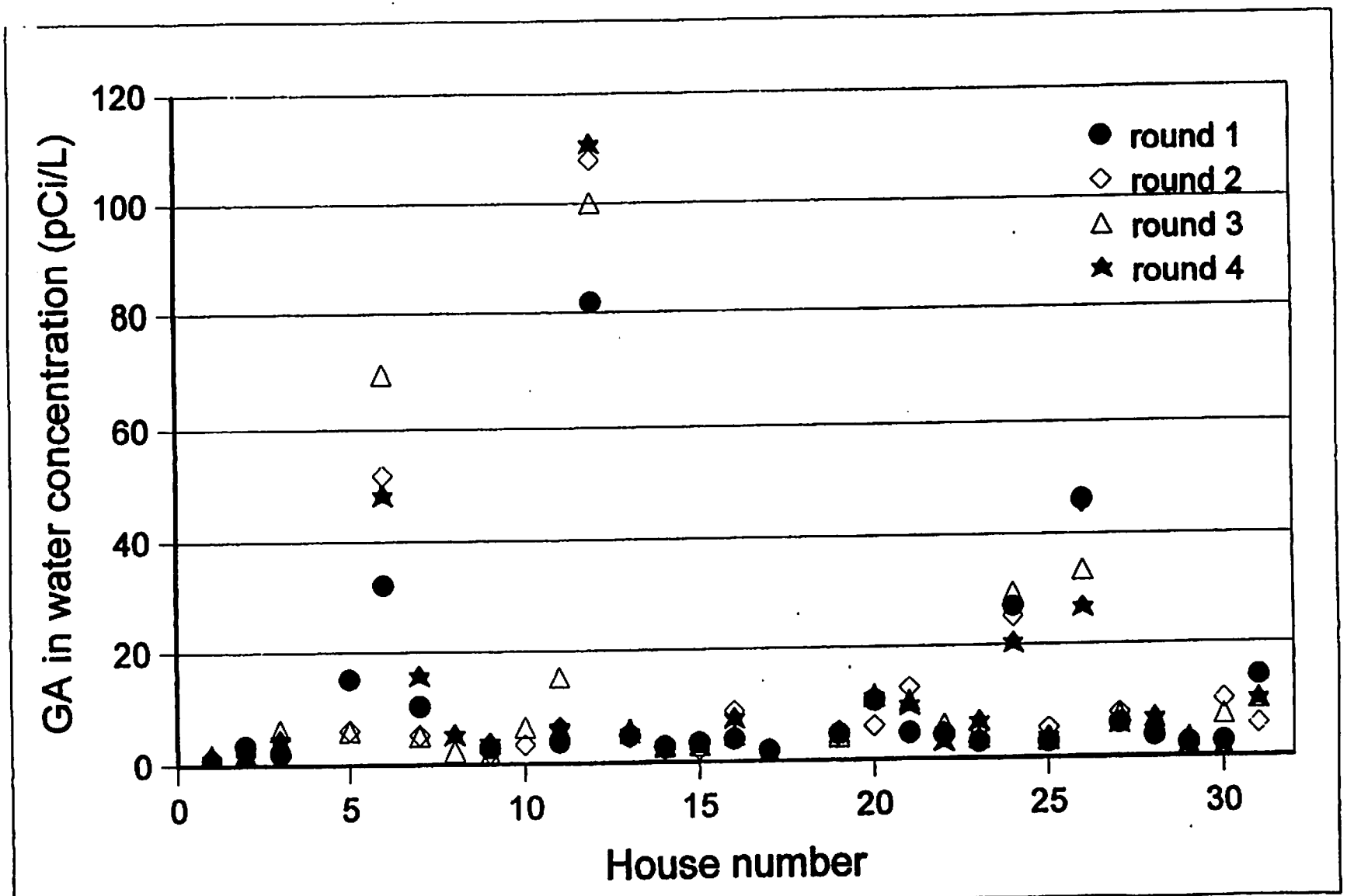

Figure 2. Waterborne gross alpha activity (pCi/l) observed in wells in Warwick during four times of the year.

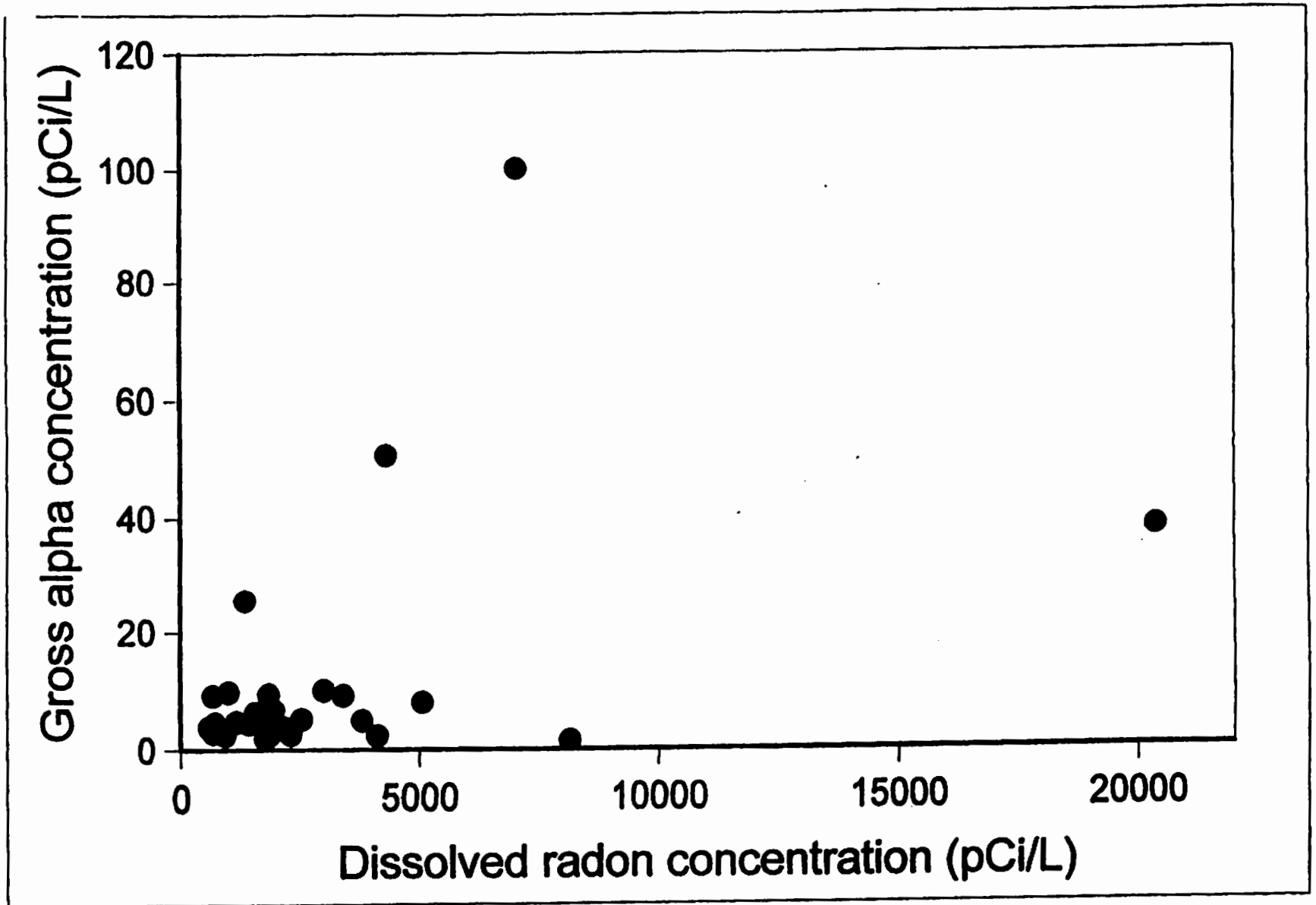

Figure 3. Correlation of dissolved radon and gross alpha concentrations observed in wells in Warwick.

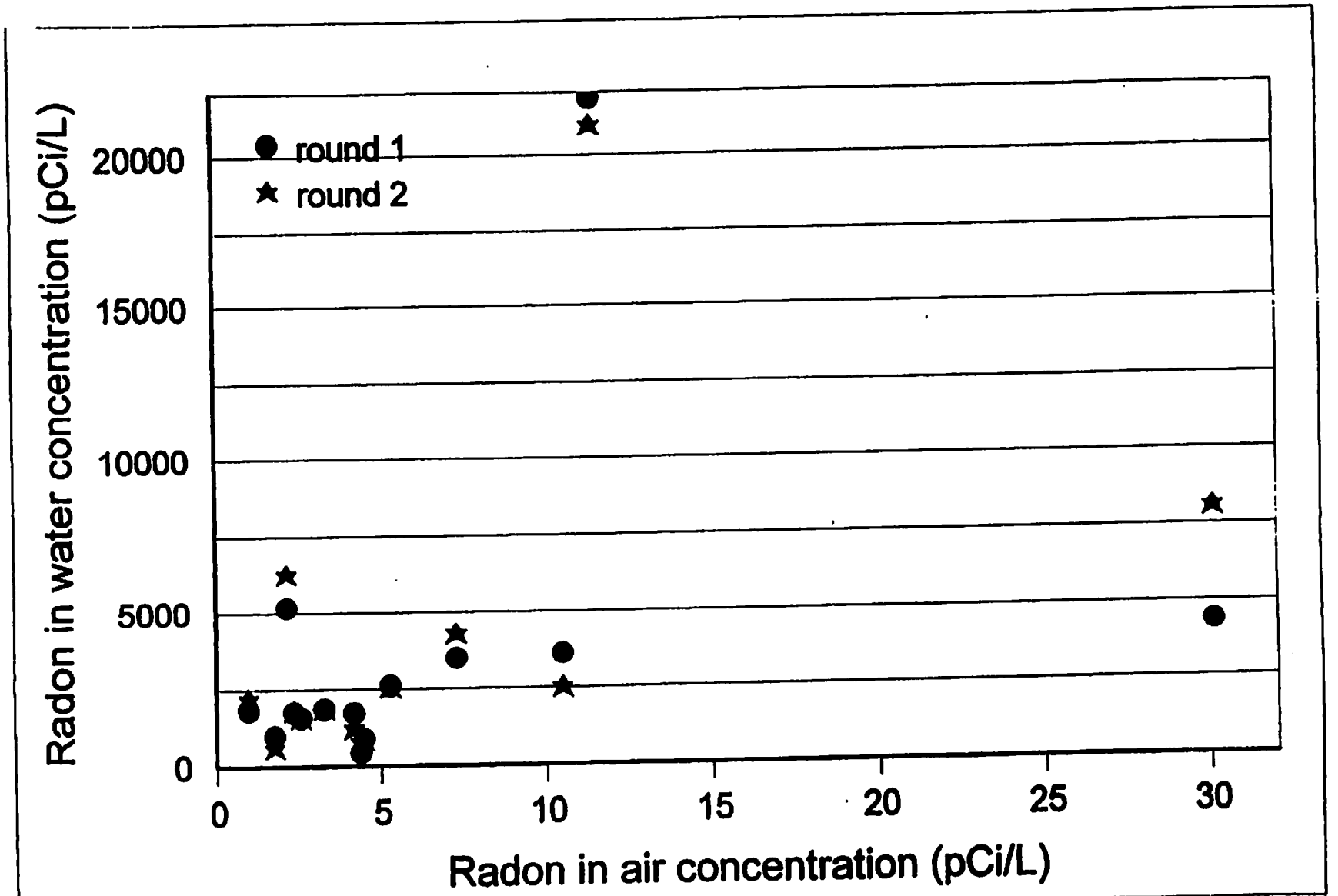

Figure 4. Correlation of dissolved and indoor radon concentrations in Warwick.