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INVESTIGATION OF RADON-222 IN SUBSURFACE WATER AS AN EARTHQUAKE PREDICTOR

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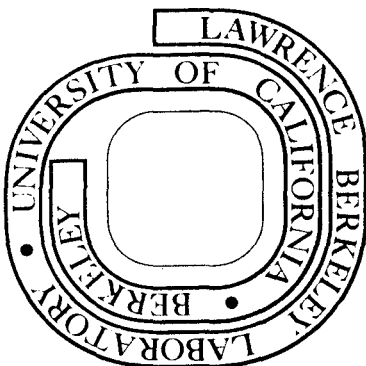
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INVESTIGATION OF RADON-222 IN SUBSURFACE WATERS  
AS AN EARTHQUAKE PREDICTOR\*

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ABSTRACT

Changes of radon-222 content of well waters in seismically active regions may provide earthquake precursor signals, according to reports of recent Chinese and Russian work. A high-precision  $\gamma$ -ray system for continuous monitoring of radon in wells and springs has been developed at the Lawrence Berkeley Laboratory, where monitoring began in April 1975, and has been extended to other sites including the San Andreas fault zone.

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## I. INTRODUCTION

Recent reports of Chinese and Russian earthquake-prediction efforts<sup>1</sup> indicate that radon-222 in subsurface waters may respond to changes in earth strain which precede earthquakes. Such changes are also reflected in some characteristics of crustal rocks within and near epicentral areas, for example: variations in the ratios of compressional and shear velocities, and apparent electrical resistivity. It is postulated that dilatancy of rocks in response to strain may enhance release of Rn-222 from the rocks into the ground water regime. Thus the onset of seismic activity may be preceded by an increase of radon content of well and spring waters in the vicinity.

We summarize here the initial progress in a collaborative effort between the University of California Lawrence Berkeley Laboratory (LBL) and the National Center for Earthquake Research of the United States Geological Survey (USGS) at Menlo Park. The goal of this work is to obtain an evaluation of the quantitative relationship between Rn-222 content of well waters and subsequent seismic activity, and if the method proves feasible, to develop compact efficient instrumentation for the job. The Rn-222 work, in turn, is part of a more comprehensive program that will also document the behavior of other chemical species in these waters.

## II. GENERAL APPROACH

Radon-222 is useful for our purpose because: 1) it is a noble gas and does not combine chemically; 2) its 3.82-day half-life is long enough for an appreciable fraction to exist for upwards of 2 weeks following birth from the parent Ra-226; and, 3) it is readily soluble in water. These three factors combine to permit transport of Rn-222 by subsurface waters over considerable intervals in time and space—thus bringing this study into the realm of practicality.

Radon-222 belongs to the U-238 series, as indicated in Fig. 1, which shows a partial decay scheme of this naturally radioactive family. Note that it is not practical to measure Rn-222 directly by  $\gamma$ -detection, but rather by detection of  $\gamma$ -rays from the two daughters Pb-214 and Bi-214. The effective half-life for ingrowth of these daughters is on the order of 40 minutes; thus our Rn-222 measurement technique must take into account a disequilibrium period that can last as long as 5 hours, which is the case for Rn-222 entering the detection volume without any daughters (Rn-222 that is either just created or freshly stripped of the daughters).

A  $\gamma$ -ray spectrometric method is used for Rn-222 assay, to provide a stable and rugged system with the capability for continuous monitoring at high sensitivity. A schematic representation of a future field installation is shown in Fig. 2. The flowing water is the source of radon to be measured, serves as a shield against external terrestrial  $\gamma$ -radiation, and provides temperature stabilization for both the detector assembly and the electronic package. Data from this equipment will be logged at the field installation and also transmitted to a central collection station.

### III. EXPERIMENTAL PROGRESS

#### A. The Continuous Monitoring Test Facility at LBL

The prototype continuous monitoring station has been in operation at LBL since April 1975, functioning as a test facility to evaluate the measurement technique and system performance. The LBL system includes an NaI(Tl) crystal detector located centrally in a tank through which radon-bearing water flows, a multichannel pulse-height analyzer (PHA) with digital gain stabilization, and peripheral units that record digital quantities derived from the PHA spectrum. A tank size of 3- to 6-ft diameter and equal height is adequate; a 3-in.-diameter by 3-in.-thick NaI(Tl) crystal in such a tank provides ample sensitivity for the relatively low-activity LBL water: 300 to 100 c/min, depending upon selection of the energy interval. A typical  $\gamma$ -ray spectrum obtained at this station is shown in Fig. 3, where prominent  $\gamma$ -peaks are assigned energy values in keV. Data is recorded as integral counts for 10-minute intervals, and shows a level that has been constant over short time intervals within the expected statistical variations. Precision of 1% or better is easily attained by summing several such intervals, permitting measurement of a few percent change in Rn-222 abundance in a small fraction of a day.

The LBL station utilizes water from a horizontal hole drilled approximately 400 ft into the hillside for slope-stabilization purposes. The drain hole provides a constant flow rate of about 1.5 gal/min of water with a Rn-222 content of  $\sim 300$  pCi/liter. The daughter products of Rn-222 (the source of the observed  $\gamma$ -radiation) are greatly depleted in water that emerges from this hole. This disequilibrium problem may be expected as a general characteristic of natural aquifers,<sup>2</sup> and must be taken into account when designing flow-through measurement systems. We have taken continuous data with two tank sizes: a 1000-gal tank, which provides a mean residence time on the order of 600-700 min, and a 55-gal tank, which has a mean residence time of 30-35 min. Equilibrium in the large tank was verified for full-flow conditions by stopping the flow and observing only the decrease in count rate expected from the 3.82-day half-life of Rn-222. Equilibrium did not exist during full-flow conditions in the small tank; there was greater than twofold increase in count rate following flow stoppage. Additional tests on discrete water samples confirmed these observations.

The Rn-222 daughters are evidently removed from the water either in transit through the aquifer or in the drain hole casing, or both. The important point to recognize is that in cases of inflow disequilibrium the residence time in any counting tank must be long compared to the 35-40 min effective half-life of the  $\gamma$ -emitters; otherwise, the (apparent) measured Rn-222 content will be flow-rate dependent. Conversely, if a short-residence-time tank is used, flow rate must be held constant if valid Rn-222 data is to be expected from experimental data.

## B. The Discrete Sampling Program

Discrete sampling of well water is an integral part of the program, ranging in application from reconnaissance studies in new areas, through long-term support in the vicinity of a continuous monitor station, to exclusive use in long-term surveillance in a study area. The broadest scope of the program includes trace-element analysis of selected samples to disclose whether other chemical precursor signals can be observed. Sample container materials must be chosen carefully so not to interfere with such measurements. Among commercially available items, polyethylene bottles are excellent for the trace-element studies; however, these containers are not impervious to Rn-222 diffusion (and subsequent loss of radon). Two types of polyethylene bottles were tested for radon-loss characteristics, as summarized in Fig. 4. Here we have plotted the measured percent of Rn-222 retained for the indicated storage times compared to the amount that should be retained in accordance with the initial measurement and a 3.82-day half-life. The upper curve, obtained from 16-ounce (~500 ml) wide-mouth bottles (NALGENE #2104), shows an acceptable loss characteristic, particularly in light of the rarity of a delay between collection and measurement of greater than 2 half-lives. This type bottle is presently in use for discrete sampling, where a sample consists of from 2 to 7 bottles (1 to 3-1/2 liters). The lower curve, obtained from the one-gallon "Cubitainer," shows an unacceptable loss characteristic for measurement of Rn-222 in water samples. The loss rate is not only large, but even relatively small variations in this rate could introduce significant errors in experimental results.

All discrete samples are transported to LBL and are assayed for Rn-222 by  $\gamma$ -ray spectrometry, where we utilize a high-sensitivity NaI(Tl) system located in the Low Background Counting Facility at the Health Physics Department. The low background counting environment is necessary for successful measurement of the small Rn-222 activities present in most water samples.

## C. The Oroville Area: Studies During an Aftershock Sequence

An earthquake of Richter magnitude ~6 occurred near Palermo in the Oroville area of the Sierra Nevada foothills (about 120 miles northeast of San Francisco) on August 1, 1975. We established a discrete sampling program for water wells on August 5, and a continuous monitoring station at one well at the Palermo Elementary School on August 27-28. We sought to take advantage here of the large number of aftershocks in an effort to establish whether our techniques could demonstrate a relationship between Rn-222 levels in well waters and seismic events, given the particular geologic terrane of the Sierra foothills and a limited selection of already existing relatively shallow wells in the epicentral area.

At present we obtain samples from six wells, four in the Sierra foothill metavolcanic bedrock formations and two in the Tertiary sedimentary deposits of the eastern margin of the

Sacramento Valley. Sampling (on at least a daily basis) is done almost entirely by the local residents, without whom this part of the study would be impossible. The discrete sampling program must continue into the period when the region is again seismically inactive, so as to establish base levels and normal variation patterns for Rn-222 content of these wells. Only then can the present data be completely evaluated to disclose which features were most probably related to seismic activity.

From mid-September onwards, intervals between aftershocks lengthened sufficiently so that we could begin to attempt correlations between changes in Rn-222 levels and seismic activity. Data from two of the bedrock wells for this period are shown in Fig. 5, where the laboratory analyses are expressed in terms of decay-corrected Rn-222 counting rates. Actual Rn-222 concentrations in terms of pCi/liter are approximately 1/3 the values shown on the graph. The occurrence times of aftershocks with Richter magnitudes greater than 2 are indicated along the base line of Fig. 5 as the tallest vertical bars.

Some preliminary comments are given here to illustrate the current understanding of the data from the discrete sampling program. The two wells (Fig. 5) exhibit marked changes in radon abundance as a function of time, but in contrasting patterns. For example, the low/high/low structures in Gilley well data from the periods Sept. 21-24 and Oct. 4-8 may be related to the aftershock sequences of Sept. 25-27 and Oct. 10-13. The high/low structures in the Prosis well data may be related to these same aftershocks, although the time scale appears to be quite different. The Gilley well data also shows a general decrease in Rn-222 levels throughout this period, in a continuation of a trend observed since earliest August, and generally following the decrease in frequency of aftershocks. Data from the Gilley and Prosis wells show less structure with passage of time during this period.

There appears to be some general correlation between aftershock activity and the radon abundances in the bedrock wells; however, the aftershocks may still be too closely spaced in time to permit distinction between precursor and response signals. On the other hand, a well drilled into the Tertiary sediments at the Palermo School (data not shown here) has a much more constant radon level, varying less than 25% during the same period.

The continuous monitoring system at the Palermo School well was set up to measure the abundance of Rn-222, using the school storage tank. Although discrete sampling had indicated the radon abundance in the water was nearly constant with time, marked variations in the apparent Rn-222 abundance were noted with the continuous system. These variations correlated closely with water usage patterns. For example, during periods of high use, the mean residence time in the storage tank was too short to achieve equilibrium between Rn-222 and daughters, and the observed count rate dropped considerably. An acoustic pickup was installed on the water system to indicate periods of pump operation, to permit possible recovery of the true radon abundance values from the data. The need for close control of water-flow patterns in such a continuous radon monitoring system was clearly demonstrated by this experience.



#### D. The Hollister Area

In March 1975, preliminary survey in the Hollister area of central California (about 100 miles south of San Francisco) disclosed the presence of Rn-222 in appreciable quantity at several wells; two wells at one site sustain considerable artesian flow during part of the year and also show relatively high Rn-222 content, approximately 800 pCi/liter. This site is in a zone of the San Andreas fault that is presently undergoing active tectonism in the form of abundant micro-and macro-earthquake activity and observable fault creep. It was chosen for the initial field station, to be occupied in the summer of 1975. However, occurrence of the earthquake near Oroville, California on August 1, 1975 caused postponement of establishment of a station in the San Andreas Fault zone until early November 1975.

The Hollister area is especially well suited for the Rn-222 study, not just because there are frequent small earthquakes, but primarily because here there are extensive instrumentation networks measuring geophysical parameters for the same purpose--sensing earthquake precursor signals. Thus the possibility exists to establish correlations between the Rn-222 method and several other methods, an opportunity not available in the Oroville area.

#### IV. DESIGN CRITERIA FOR A CONTINUOUS MONITOR FIELD STATION

The continuous monitoring system at LBL was assembled from general purpose laboratory equipment, as was the apparatus set up at the Palermo Elementary School in the Oroville area. The latter system was recently moved to the Hollister area, where an air-conditioned trailer houses the electronic units. Design criteria for the specialized field units are being formulated and refined in light of experience accrued at these two fully instrumented field stations.

Present concepts for the signal-processing path include an analog-to-digital converter (ADC) and digital gain stabilization, to insure that the energy calibration cannot drift, short of some catastrophic failure. The most intense peaks in the equilibrium Rn-222  $\gamma$ -ray spectrum are clearly resolved with the NaI(Tl) spectrometer system, as is shown in Fig. 3, and are thus suitable for gain stabilization. One can also select clearly identifiable energy bands from the spectrum, which provide both the Rn-222 data and verification of correct system operation. For example, counting rates of two intervals within the Rn-222 spectrum (when corrected for ambient background) should maintain a constant ratio regardless of Rn-222 levels, and this constancy would indicate that the electronic equipment and detector were functioning properly. A third quantity, the number of events in the gain-stabilization interval, should track the above two quantities; any abrupt changes in these ratios would indicate a malfunction. The recording of all events above about 4000 keV, a gross measurement of the cosmic-ray intensity, would provide continuous verification of detector efficiency; no natural terrestrial  $\gamma$ -rays are energetic enough to register above this threshold, and the cosmic-ray intensity is nearly constant with time.

These digital data quantities, recorded locally or sent to a central collection station, would be defined by digitally determined upper and lower bounds derived from the gain-stabilized "spectrum." System reliability and data credibility are thereby greatly improved. Microprocessor technology is being considered for processing and transmitting of station data, and for controlling station operation.

### SUMMARY

The intent of this work is to develop a compact high-precision real-time continuous monitor system for field measurement of Rn-222 in subsurface waters, an instrument that could serve as a prototype for a network of such stations. Data from a group of these stations could then be coordinated with other geophysical information related to earthquake precursors, to be used in earthquake prediction.

Although the work reported here focuses on the  $\gamma$ -ray spectrometric method, application of  $\alpha$ -particle detection to this purpose is also being vigorously pursued, especially by the USGS side of the collaboration. A progress report on the  $\alpha$ -particle work will be given by one of the authors (Chi-Yu King) at the meeting of the American Geophysical Union in San Francisco on December 8, 1975.

Initial progress in the  $\gamma$ -ray detection method includes the following items. We have verified the major design features of a field station that performs continuous high-precision Rn-222 monitoring in water. We have established a long-term locally supported discrete well-water sampling program in the Oroville area during the period of aftershock activity, to investigate the link between Rn-222 levels and the ongoing seismic activity. Preliminary results indicate some correlation between seismic activity and radon abundance patterns in the Oroville area, but final evaluation of the data must await return of normal seismic inactivity.

### ACKNOWLEDGMENTS

We express deep appreciation to the following LBL personnel: Dr. Earl K. Hyde, Deputy Director, for his continuing support of the program; Dr. Ralph H. Thomas, Head of Health Physics, within whose department all discrete sample measurements are done; to Anna Henson, for continuing assistance in data analysis; and to James E. Arnold, for his invaluable electronic support work.

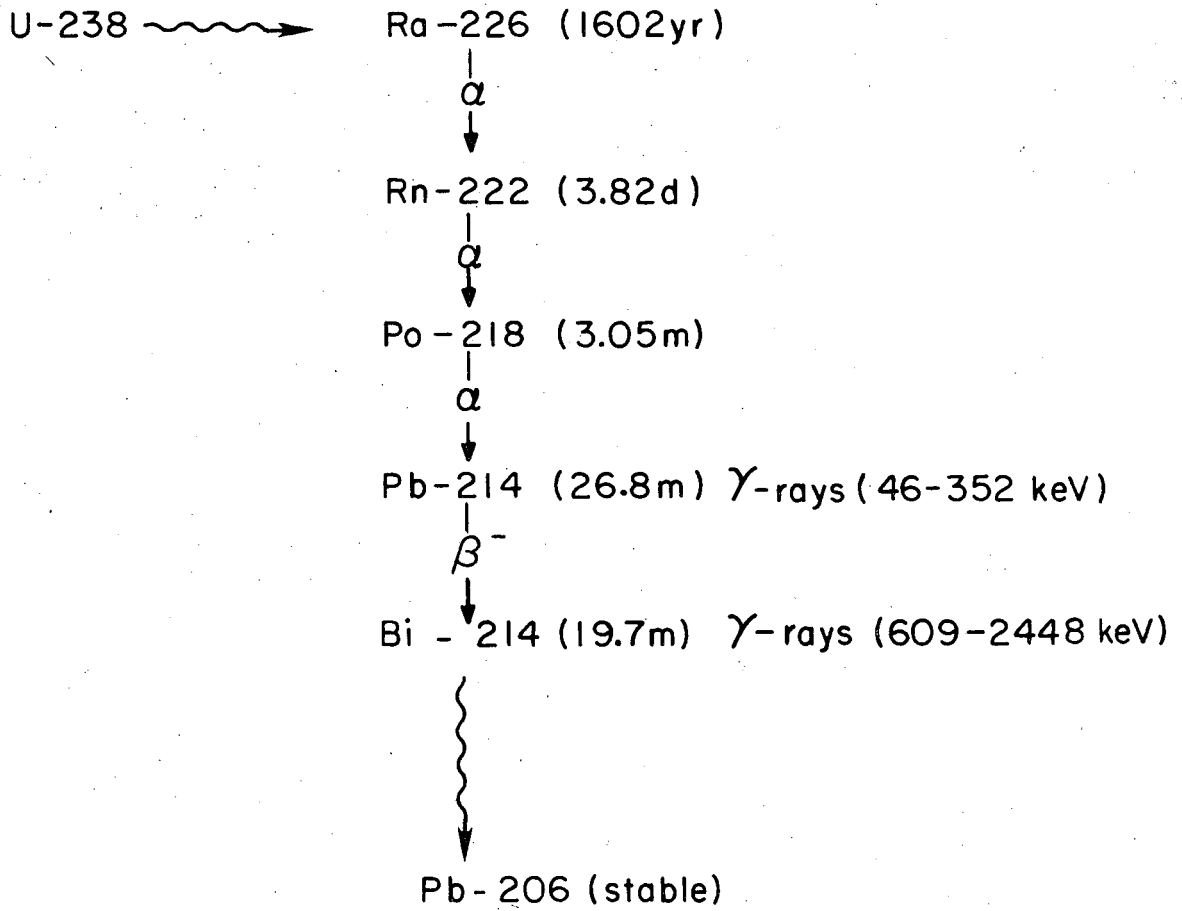
We are indebted to Dr. C. B. Raleigh, of U.S. Geological Survey National Center for Earthquake Research, for his initial organization of the collaborative program, and for continued encouragement and advice.

We are also most appreciative for the interest and assistance given by the administration, teaching staff, and students at the Palermo Elementary School, and to the owners of the other

five wells in the vicinity; H. Gilley, Jr., W. Mooney, P. Norman, G. Prorise, and D. Webb. Without their continued participation, the discrete sampling program would be impossible.

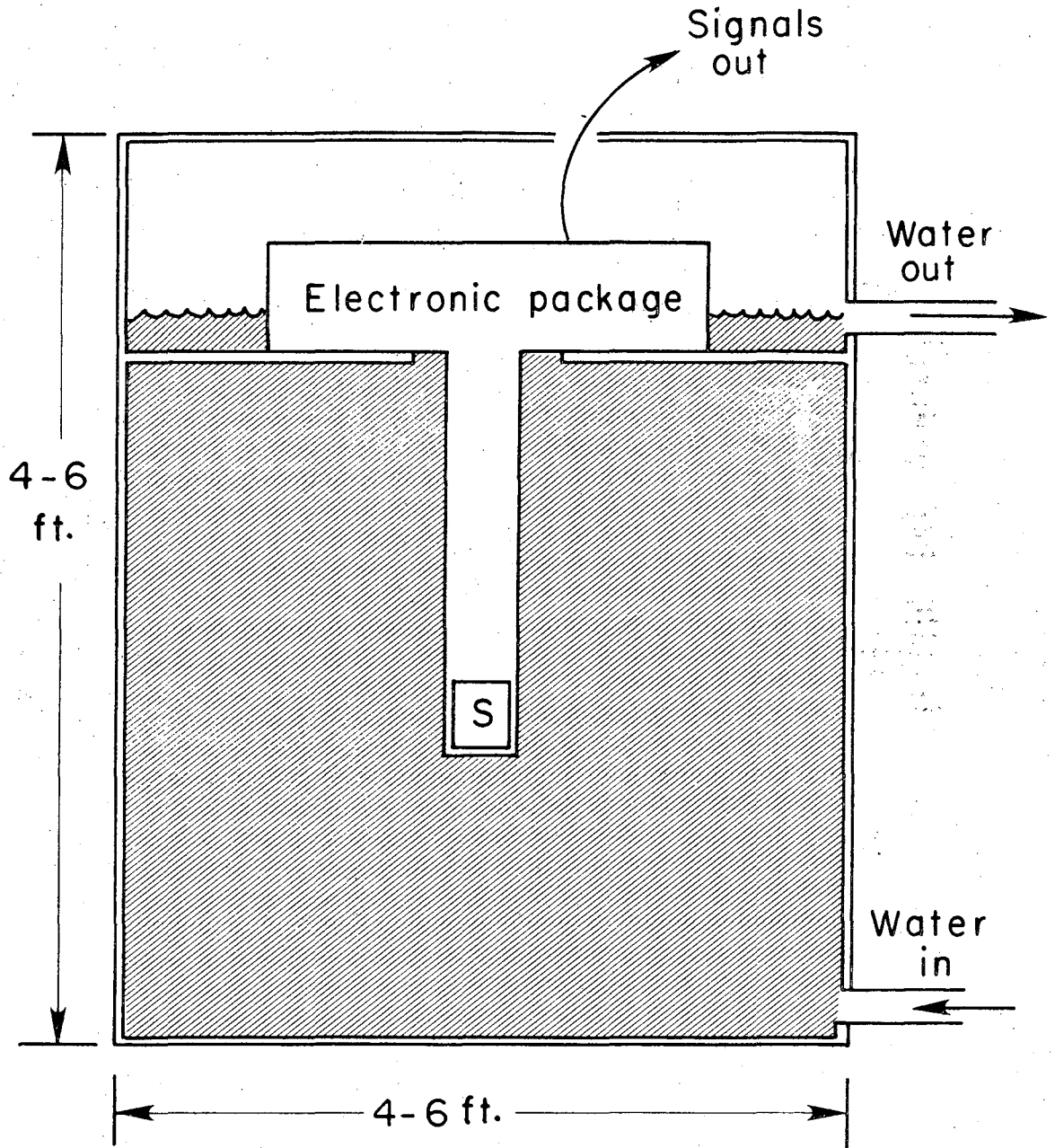
#### REFERENCES

1. Frank Press, "Earthquake Prediction," *Scientific American*, vol. 232, No. 5, pp. 14-23, 1975.
2. Allan B. Tanner, "Radon Migration in the Ground, A Review," and "Physical and Chemical Controls on Distribution of Radium-226 and Radon-222 in Ground Water Near Salt Lake City, Utah," Chaps. 9 and 13 in The Natural Radiation Environment, edited by J. A. S. Adams and W. M. Lowder (University of Chicago Press, 1964).



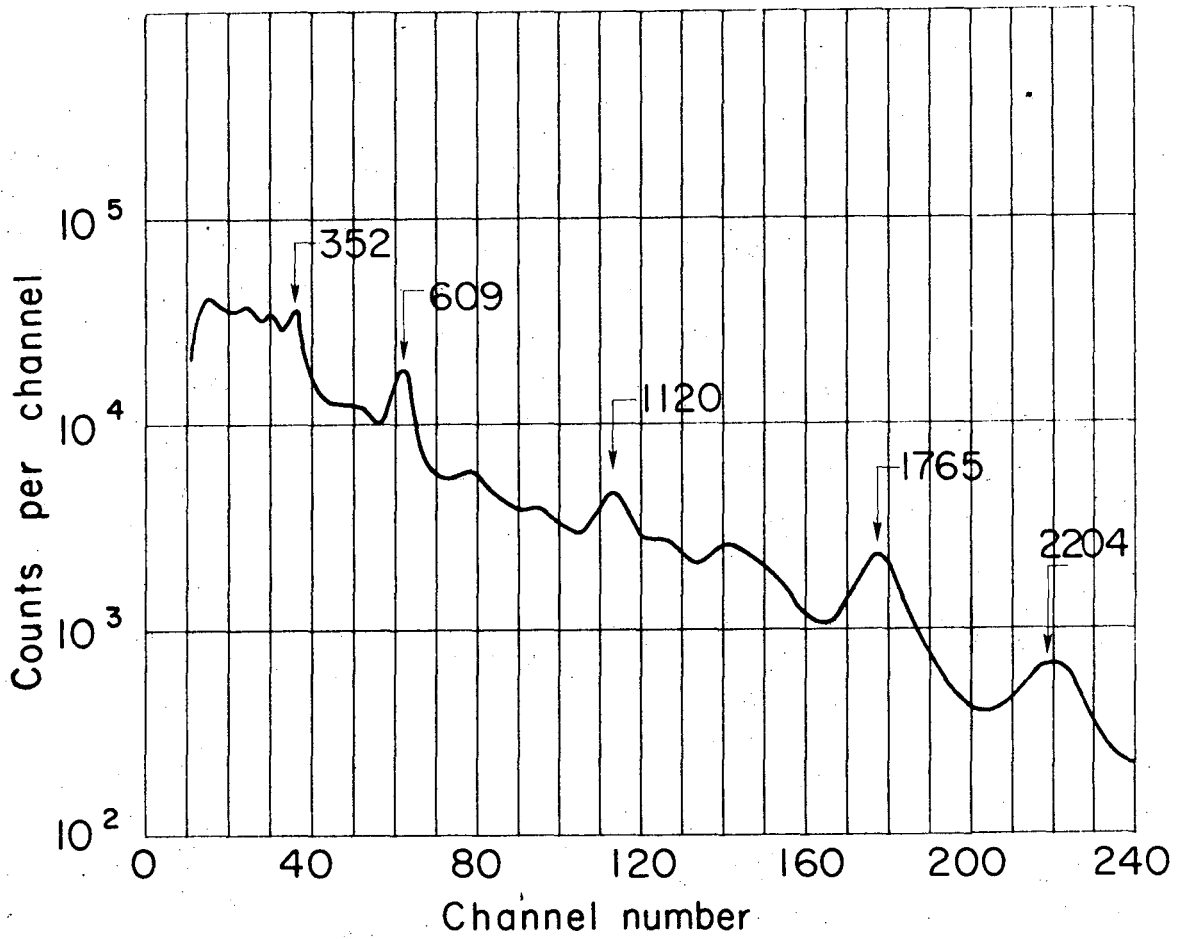
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Fig. 1. Partial decay scheme of the U-238 series.



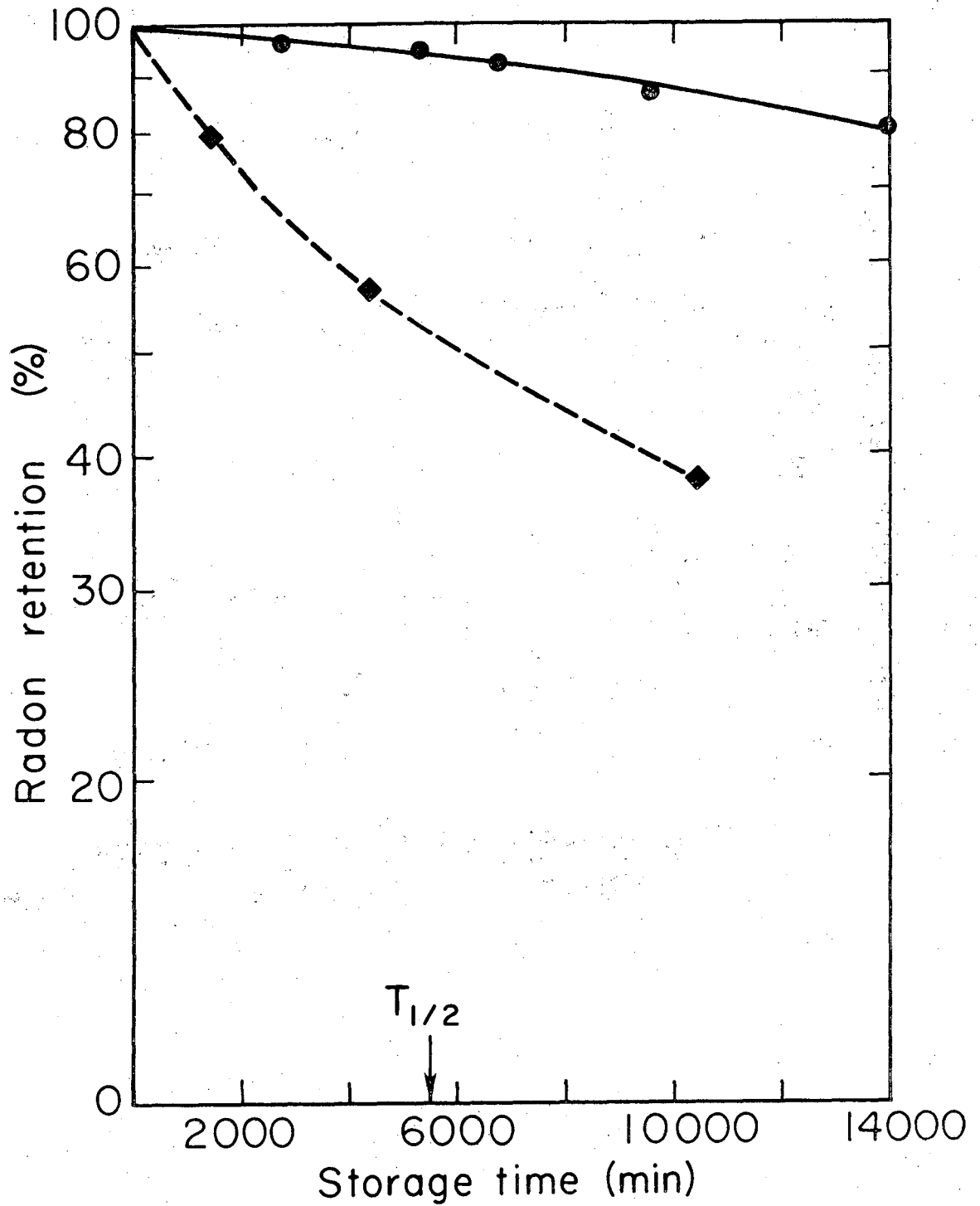
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Fig. 2. Schematic layout of field facility for Rn-222 measurement in water by  $\gamma$ -ray spectrometry.



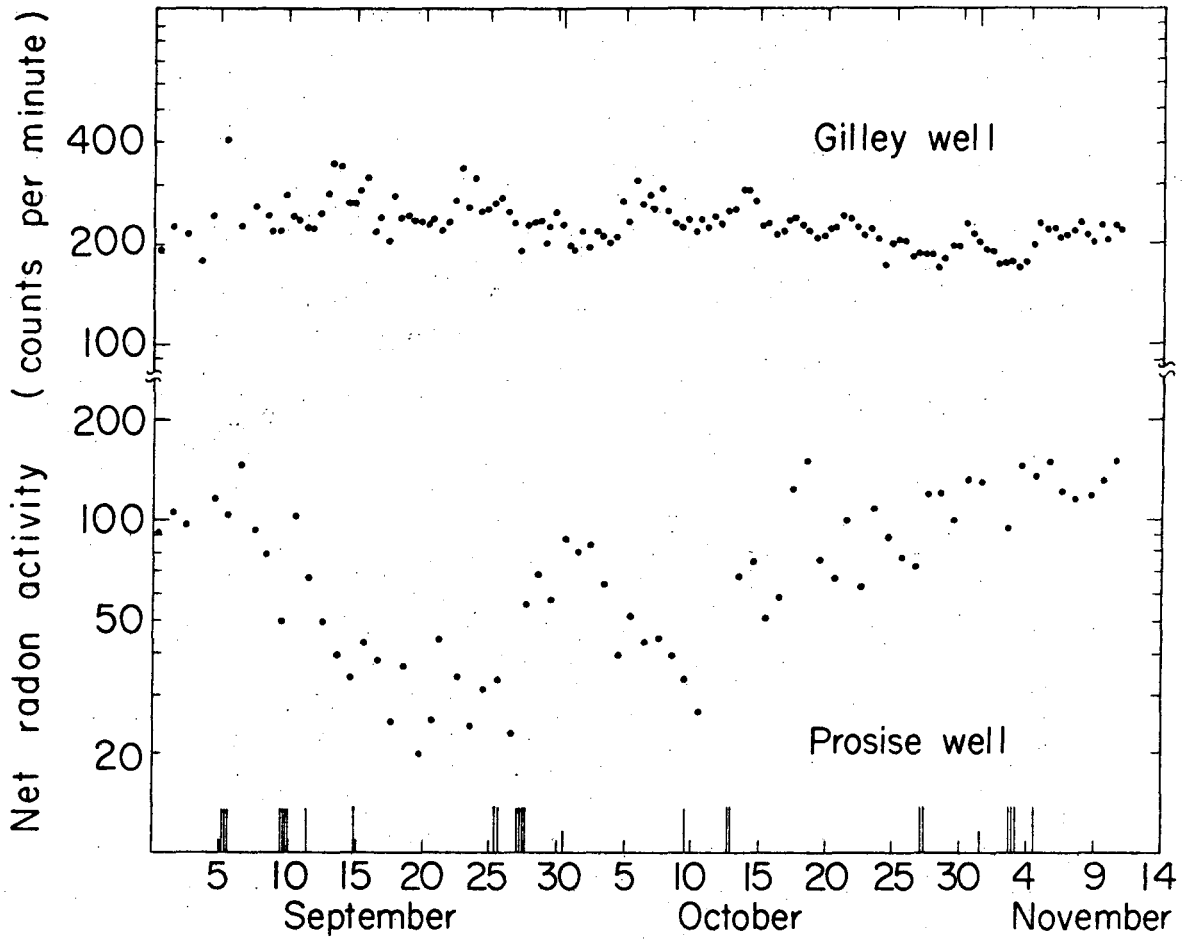
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Fig. 3. Gamma-ray spectrum obtained at the LBL continuous monitoring test facility.



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Fig. 4: Retention of Rn-222 in water for two types of polyethylene sampling containers.



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Fig. 5. Results of Rn-222 measurements in the discrete sampling program from two bedrock wells in the Oroville area.



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