Improvements in the Measurement of Rn-222 in Natural Waters•

Bill Burnett, Taylor Nelson, Reide Corbett

Environmental Radioactivity Measurement Facility, Department of Oceanography, Florida State University, Tallahassee, FL 32306; email: burnett@ocean.fsu.edu

Larry Robinson and Judith Weaver

Environmental Sciences Institute, Florida A&M University, Tallahassee, FL 32307

J.E. McKisson

Radiation Technologies, Inc., 13101 Rachael Blvd., P.O. Box 459, Alachua, FL

Derek Lane-Smith

Durridge Company, 7 Railroad Ave., Suite D, Bedford, MA 01730

The direct discharge of groundwater into coastal and other surface waters is a common process which could lead to environmental deterioration in cases involving contaminated groundwater. Past studies have shown that areas of significant discharge via submarine springs or seeps may be located using ^{222}Rn as a natural tracer. Unfortunately, detailed studies have been limited by the logistics and time-consuming nature of sampling and analyzing water samples in the traditional manner. We have begun the process of developing in situ and continuous radon monitors that will be capable of producing automatic measurements of ^{222}Rn in surface waters. This will provide a significant advantage in the assessment of groundwater seepage sites into lakes, rivers, and coastal areas.

We describe here three approaches for the quantitative assay of ^{222}Rn in natural waters for levels down to approximately 0.1 dpm/L (~0.5 pCi/L or 0.002 Bq/L), the approximate background concentration of radon in coastal ocean water unaffected by submarine groundwater discharge. The three approaches include: (1) a radon emanation system that sparges ^{222}Rn directly from water samples; (2) an "active" processing system that would bring a stream of water to an equilibrator where it is mixed with air that recirculates through another loop an in-line atmospheric radon monitor (RAD-7); and (3) a submersible "passive" unit which will provide continuous radon measurements at one location over long periods of time.

The **radon emanation system** has been in operation in our laboratory for several years and represents an advancement over the standard EPA-type system for radium analysis which collects Ra-226 as a coprecipitate with $BaSO_4$, redissolves the precipitate in EDTA, and introduces the solution into a micro de-emanation system. Our approach has been to design a gas line that allows us to sparg several samples directly with helium, remove moisture and CO_2 via gas purification tubes, collect the Rn on liquid nitrogen cold traps, and then transfer the gas into custom-designed alpha scintillation cells. The system works very well for both radon and radium and can be built fairly inexpensively. We currently have sufficient ports

---

• For presentation at the 44th Annual Conference on Bioassay, Analytical, and Environmental Radiochemistry, Albuquerque, New Mexico, November 15-20, 1998.

and counters to allow us to process up to ten samples simultaneously. Unfortunately, for some of our needs, even that level of sample production is insufficient. Thus, the desire for some type of continuous measurement device.

The **continuous radon monitor** is based on a modification of a standard unit for continuous measurement of atmospheric radon called the RAD-7 (manufactured by Durridge, Bedford, MA). This monitor is based on the electrostatic attraction of radon daughter products to a semiconductor detector. Thus, the unit has energy discrimination and very low backgrounds allowing a detection limit for ^{222}Rn in water of approximately 0.01 pCi/L (depends on temperature). For continuous measurements of water, an exchanger will mix a stream of water with air that recirculates through the RAD-7.

Experiments were run at Durridge using a prototype exchanger and water from the town of Bedford’s water supply (known to be fairly high in radon). The measured radon concentration is that of the air circulated to the RAD7. The radon concentration in the water can then be calculated using the equilibrium coefficient given by:

$$a' = 0.105 + 0.405 * \exp(-0.0502*T)$$ where T is the temperature in degrees C.

This relationship is from Von Fritz Weigel, "Radon", Chemiker Zeitung 102:287 (1978).

During the experiments at Durridge, the water temperatures stayed within 0.5 degrees of 19 degrees C. So a' was approximately 0.261 for these tests. So one may simply multiply the radon-in-air values by a' to obtain the radon-in-water concentrations.

The water supply in the town of Bedford has a radon concentration around 25 pCi/L. Water for the area is typically obtained from reservoirs, so this value is reasonable. Sometimes, however, they use artesian wells, and Durridge reports, on one occasion, observing values close to 1,000 pCi/L in the water.

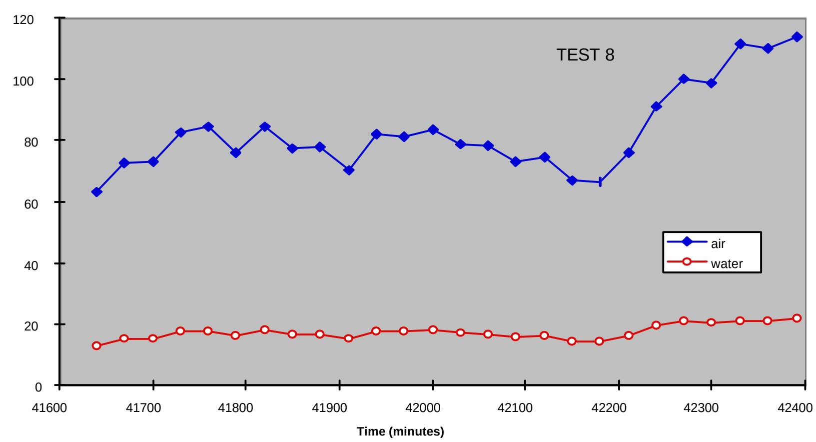

Figure 1 Long-term test of a prototype exchanger system with a modified RAD-7. The closed symbols represent the radon-in-air concentration while the open symbols represent radon-in-water (assuming equilibrium).

The "Test 8" data set (**Fig. 1**) is a long-term measurement (800 minutes) which was started after the system had already been running for several hours. There is good evidence for equilibration between the re-circulated air and the water stream. Interestingly, the plot shows a distinct rapid increase in concentration, which occurred on Monday morning. We speculate that this may be associated with the sudden increase in use of town water in the morning. That could represent the time when they add well water to the supply. Well water is usually much higher in ^{222}Rn concentration.

The **submersible radon monitor** (under construction) will consist of a gas-filled chamber separated from the ambient water by a hydrophobic, microporous membrane (**Fig. 2**). Radon, without accompanying water, will diffuse into the chamber. Following radioactive decay, the charged daughter projects (^{218}Po^{+} and ^{214}Po^{+}) will be electrostatically precipitated onto an active detector surface. Entry of the subsequent alpha particles into the detector will produce a pulse which will be measured and displayed in a multi-channel analysis system.

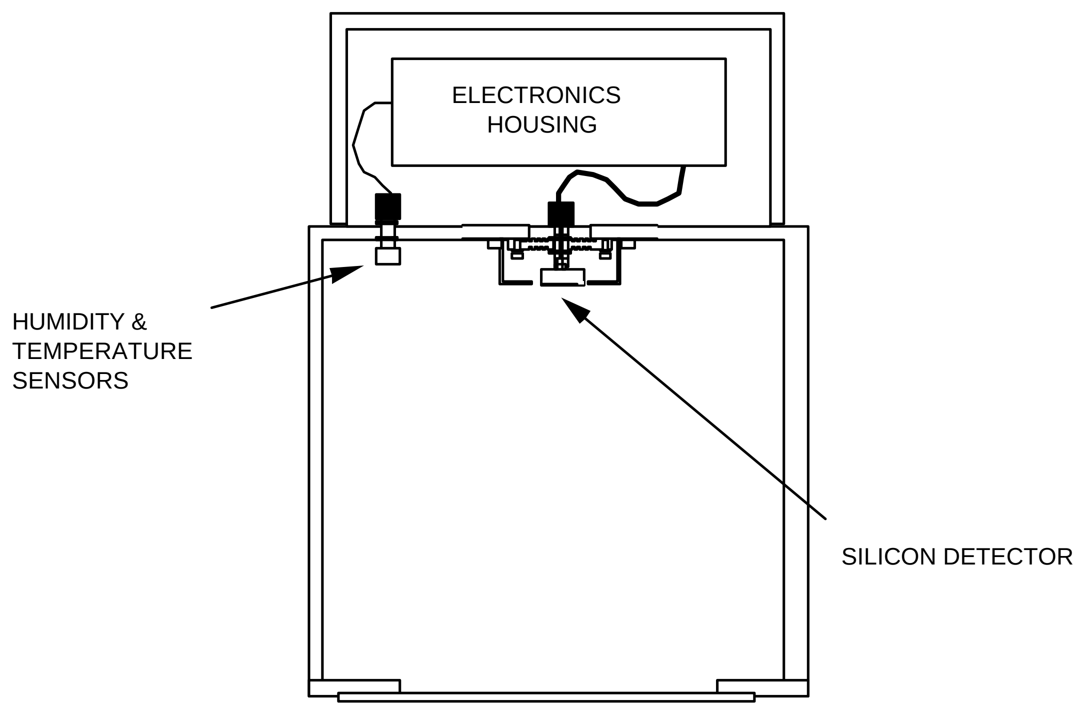

Figure 2. Suggested layout of humidity and temperature sensors with respect to alpha particle detector. The cup also shields the detector element from any stray fields associated with the humidity sensor.

The electronics package has been completed by RTI and an atmospheric test was run in the RTI shop overnight with the detector exposed to ambient air in an open-top aluminum enclosure (Faraday shield). The detector was mounted in its Teflon mount and then placed on a Teflon sheet to minimize leakage and was then exposed to ambient (laboratory) air in an open-top aluminum enclosure. The test conditions were as follows: electrostatic voltage -2500 V; temperature/humidity sensors disabled; output signal buffered; and cable length 20

m. Data were acquired with an Aptec 5000 Series MCA with bipolar 4.0 μs shaping into 4096 channels. A spectrum acquired over a 12-hour period is depicted in **Figure 3**.

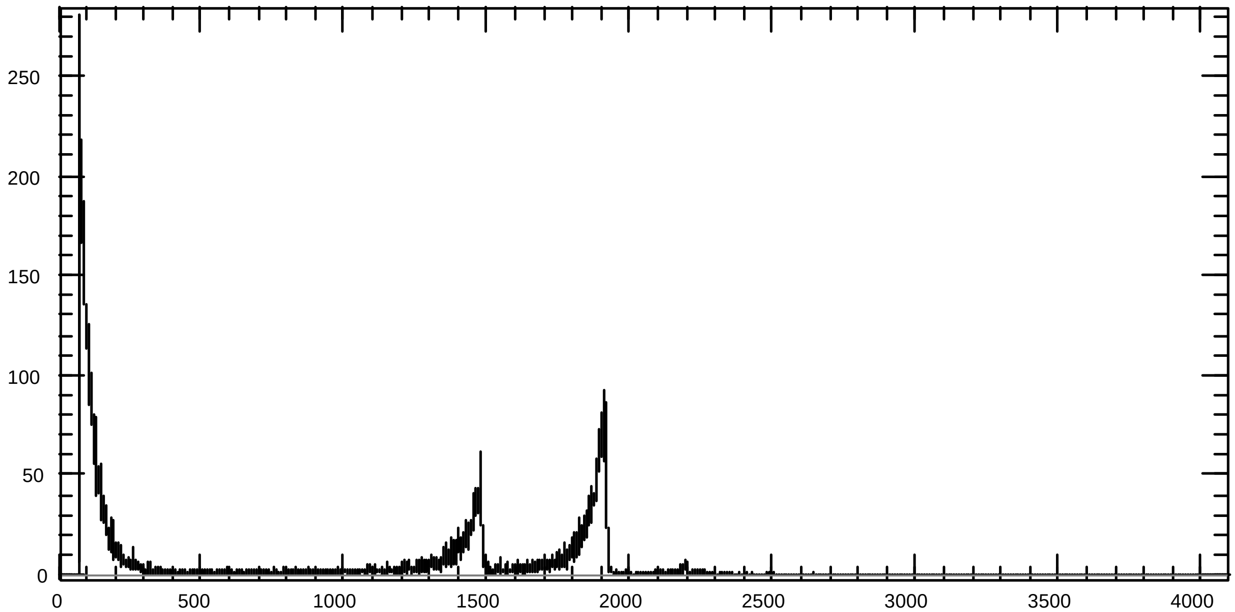

Figure 3 Spectrum (x-axis: channel number; y-axis: counts) accumulated overnight at RTI with the detector exposed to ambient air. The two main peaks are due to the radon daughters ^{218}Po and ^{214}Po.

Assuming we were looking at the 6.0 MeV and 7.69 MeV from ^{218}Po and ^{214}Po as the two principal peaks, the small peak to the right seems to be about 8.75 MeV. This is probably ^{212}Po (8.78 MeV) which is in the Thoron decay chain. The resolution of the detector (as measured by FWHM of both peaks) was about 0.125 MeV, depending on the software used. Much of the commercial MCA software expects relatively symmetrical peaks as from a gamma spectrum, rather than the "tailing" peaks common in alpha spectrometry, and therefore may over-estimate the FWHM.

The detector housing is currently being constructed in the FSU Department of Oceanography machine shop and we plan to begin testing the unit in an aqueous radon test tank which has been built at Florida A&M University. The test tank has a known amount of radon produced by "radon generators", cation exchange resin impregnated with ^{226}Ra and isolated from the water by a plastic membrane. This arrangement allows ^{222}Rn to diffuse into the water while retaining the radium on the resin.

This research is being supported by grant DSWA01-97-1-0022 to Florida State University from DOD's Defense Special Weapons Agency.