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Radon Solubility in Water as a Function of Salinity and Temperature

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THE FLORIDA STATE UNIVERSITY

COLLEGE OF ARTS AND SCIENCES

RADON SOLUBILITY IN WATER AS A FUNCTION OF SALINITY AND TEMPERATURE

By

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"Thinking: the talking of the soul to itself." – Plato

Going back to school while still working full time has been one of the most challenging decisions in my life, but I made it through all of this in no small part to the support of everyone in my life. First I would like to thank my family, who has been there with my the entire time and encouraged me to keep on working towards my goals no matter how difficult it may be. To my mom, dad and sisters who have helped me edit and review everything, even when they barely understand what I am talking about at times. To all of my friends who have acted as my sounding board. I want to thank my advisor, Dr. Bill Burnett who has guided me from original inception, to final writing. Who has tirelessly answered my multiple questions and addressed the many problems that have arisen during the course of experimentation. I would like to thank my committee, for taking the time and energy required for all of this to be completed. I want to thank the good people at Duridge Company, without their detector none of this research would have been possible. Last but not least, I want to thank everyone in the Oceanography department who has helped guide me and made sure I didn't sway off the path.

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ABSTRACT

Radon is an important natural tracer for certain aquatic environmental studies. New methods for rapidly determining submarine groundwater discharge (SGD) into surface waters in the coastal zone rely on the accuracy of the air/water partitioning coefficient for doing radon mapping of coastal waters. The original research that determined the radon partitioning coefficient was conducted in 1916 and more recent published analysis of these data only accounted for temperature changes in fresh water.

My research was designed to: 1) determine the partition coefficient by accurately measuring the gas and water phase concentrations of ²²²Rn when the radon is in equilibrium between both phases; 2) minimize the error due to multiple analytical system calibrations; and 3) calculate the partition coefficient at different temperatures and salinities.

I focused on 2 different experimental setups, one designed to directly measure the radon concentration of the gas phase and the dissolved concentration in the liquid phase (known as the direct measurement or volume independent method), and the other was hoped to be a rapid measurement system that only required a continuous measurement of the gas phase to determine the partitioning coefficient at different temperatures (the indirect measurement or volume dependent method).

I combined my research results with parallel research experiments being conducted in Leipzig, Germany and together we redefined the air/water partitioning coefficient of radon gas with respect to both salinity and temperature. By analyzing our combined results, we developed a new set parameters $a_1 - b_3$ for use in the Weiss Equation, which could then be used to determine the partitioning coefficient of radon at different salinities and temperatures. Our results showed that if salinity effects are neglected in warm, relatively low salinity waters (e.g.,

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coastal waters from the Gulf of Mexico), there was a slight overestimation of the radon concentration (up to 10%), but in colder, higher salinity waters, this overestimation could exceed 20%.

CHAPTER ONE INTRODUCTION

1.1 Historical Background

Historically, studies involving river discharge rates, submarine groundwater discharge (SGD) and other methods of fluid transport have utilized some form of tracer dye (visual, radiological, etc.), making these studies potentially harmful to the environment (Field et al. 1995) and requiring long periods of time while the artificial tracer is transported to the study site. Radon is normally only found at high concentrations in groundwater, which quickly degasses when exposed to surface water supplies (Cothern and Rebers 1990). Radium concentrations in the atmosphere are extremely small, so all radon in air is considered to originate from the ground and degasses based upon the permeability of the continental material (NCRP 1975). The estimated world radon release is 2.5-3 GCi/yr, which averages to 100 pCi/m³ (3.7 Bq/m³) in the northern hemisphere (Harley 1975). By utilizing the natural radon component of groundwater, and its relative depletion of radon in surface waters, discharge studies can be conducted without injection of potentially harmful tracer into bodies of water.

Traditional methods of measuring radon activities require collection of discrete water samples, bringing the samples back to the laboratory and using bulky liquid scintillation counters (Kitto 1994) or sparging water samples with inert helium-4, collecting the radon inside a cryogenic trap and then warming the cold trap back to room temperature to transfer the radon into a Lucas Cell (ZnS scintillation cell) for counting (Kitto et al. 1995). These setups require large amounts of time (> 5 hours per experimental run) and have to be conducted in a laboratory, making rapid measurements in the field (e.g., coastal mapping) difficult.

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The partitioning coefficient is the ratio at equilibrium of the dissolved gas concentration in a medium (as water) to the concentration remaining in the gas phase. By circulating the gas in a closed loop through a liquid spray chamber and into a radon-in-air monitor, the air equivalent radon concentration of the liquid can be determined in as little as 5-15 minutes once equilibrium is established (Burnett et al. 2001). During such field studies, temperature measurements are used to calculate the partitioning between radon in the air and water phases. By using these values, along with the air equivalent radon concentration, it becomes possible to accurately determine the radon concentration dissolved in the liquid phase. Unfortunately, these values have only been well established for pure water.

When the partitioning coefficient of a gas-liquid system is needed, one needs to equilibrate the gas with the liquid system and then measure the concentration in both phases. By utilizing this coefficient, it becomes easier to measure the concentration in a phase, like water where it is difficult to directly measure radon, by using the concentration in the gas phase, that is easier to measure and multiplying it by the partitioning coefficient. For example, one can more easily measure radon-in-air by utilizing a method that allows the systems to reach equilibrium (like a spray chamber). It then becomes easier and faster to precisely measure the radon concentration dissolved in the liquid media by utilizing the partitioning coefficient of radon gas between air and water (Lane-Smith et al. 2002).

1.2 Empirical Approaches

1.2.1 Weigel Equation

In 1978, Weigel published an empirical equation that relates the partition coefficient, k, in pure water to temperature (°C) that led to what is now referred to as the Weigel equation,

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K =
$$\frac{^{222}Rn(l)}{^{222}Rn(g)}$$
 = .105 + 0.405 * e^{-.05027t} (Equation 1)

This equation calculates the partition coefficient of 222 Rn between pure water and air by assuming a constant salinity value of 0 ‰ and *t* as the temperature in °C (Fig. 1). The original dataset, Weigel used to create equation 1, was published by Meyer and Schweidler (1916).

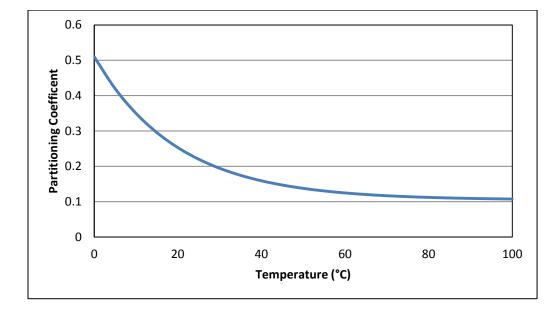


Figure 1. Exponential Plot of the Weigel Equation. The theoretical radon partitioning coefficient of pure water from 0 to 100 °C. Calculated from Eq. 1.

1.2.2 Kofler Dataset

In 1913, Kofler reported experimental data points for radon solubility in saline water which was later published by Clever (1979). Based upon the Kofler data set, at normal ocean salinity (35 ‰) and an average coastal temperature of 18 °C, the partitioning coefficient of radon would equal 0.280. When compared to the Weigel equation at the same temperature the coefficient would be 0.269. Thus, there is a significant difference in the coefficients and the resulting radon activity. The Kofler data set for radon solubility as a function of salinity may not have been scrutinized to the level that the pure water values were (Schubert et al. 2012).

1.2.3 Weiss Equation

A general approach to creating a temperature and salinity based partitioning coefficient utilizes an equation derived by Weiss (equation 2) for other noble gases, including He, Ne, Ar and Kr (Weiss 1970; Weiss 1971; Weiss et al 1978).

$$ln\beta = a_1 + a_2 \left(\frac{100}{T}\right) + a_3 \ln\left(\frac{T}{100}\right) + S\left\{b_1 + b_2 \left(\frac{T}{100}\right) + b_3 \left(\frac{T}{100}\right)^2\right\}$$
(Equation 2)

The Weiss equation calculates the natural log of the Bunsen coefficient, where S is the salinity and *T* is the temperature in degrees Kelvin. The α and β coefficients can be determined experimentally. The partitioning coefficient can be calculated by utilizing this equation,

$$K_{w/air} = \beta * \frac{T}{273.15}$$
 (Equation 3)

With the Bunsen coefficient calculated in equation 2, to create a temperature and salinity based partitioning coefficient. Before our work, in collaboration with Michael Schubert at the Helmholtz Centre for Environmental Research (UFZ) in Leipzig, Germany, none of the *a1* to *b3* parameters were available for radon gas. We thus decided to set up a series of experiments to determine these coefficients.

1.2.4 Extrapolation of k' From Other Noble Gases

To date, evaluating the partition coefficient for radon as a function of salinity has been based upon extrapolating the solubility of other noble gases: He, Ne, Ar and Xe, based upon their respective atomic weights. Using the concentrations of several other noble gases in dry air and reported concentrations in seawater given in Pilson (1998) Table 5.1 and appendices D.5 and D.6, one can extrapolate the partition coefficient for radon at different temperatures and salinities. For example, by utilizing these extrapolated values at a temperature of 20 °C and a salinity of 30 ‰ (Fig. 2) and comparing the derived value (0.231) to the value given by the Weigel equation for pure water at 20 $^{\circ}$ C (0.253), a difference of about 9% is calculated between the two values. Presumably, this difference is due to the salinity effect.

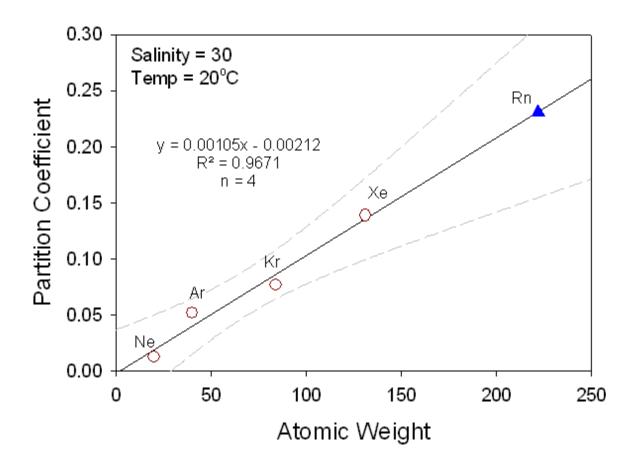


Figure 2. Pilson Extrapolation. Plotted values from Table 5.1 and appendices D.5 and D.6 from Pilson 1998. By plotting the partitioning coefficient vs the atomic weight of Ne, Ar, Kr and Xe at a salinity of 30 ‰ at 20 °C, the partitioning coefficient of radon (atomic weight 222) was theoretically determined to be 0.231. Dashed lines represent the 95% confidence limits of the regression line.

Many field surveys have been conducted in estuarine and coastal environments by calculating the water radon concentration with a RAD-AQUA (Durridge Company, Inc., Billerica, MA) spray chamber utilizing the air-equivalent radon values, corrected with the Weigel equation, to determine the water concentration (Dulaiova et al. 2005). Comparisons between the RAD-AQUA approach with traditional sample collection and laboratory analysis often showed differences less than about 10%. This led many researchers to conclude that the salinity effect was small and as such has been ignored for many years.

It was later discovered that the reasonably good correspondence between the RAD-AQUA approach, using the partition coefficient for pure water, and traditional laboratory measurements may only be the case for warm, less saline waters, such as Florida and the Great Barrier Reef (Australia), where many of the intercomparisons were performed (Lambert and Burnett 2003; Stieglitz et al. 2010).

Based on results that will be shown here, one would expect a greater difference in cooler and more saline environments, such as those found in high latitudes or in highly saline environments such as the Dead Sea. To reconcile these differences, and to clarify the role of salinity in the air-water partition coefficient for radon, I designed an experiment to directly measure both the air radon concentration and the dissolved radon concentration in a sealed container, containing water of known temperature and salinity, once it reached chemical equilibrium across the air-water interface.

In order to test the partitioning coefficient under standard atmospheric conditions, the total radon in the system could not exceed a level where the liquid phase was completely saturated with radon. This differs compared to regular gas saturation due to the fact that radon, as a noble gas, can only be oxidized by halogen fluorides (Stein 1970). Due to the inability of radon to be oxidized by water, radon cannot form a precipitate to maintain a constant concentration in the liquid and gas phase past full saturation. When additional radon is introduced above the saturation point, the excess radon accumulates in the gas phase, unless the pressure and/or the internal temperature of the system increases. This is not an issue with

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environmental testing due to the naturally low abundance of radon in the system, but must be accounted for and controlled when applied to laboratory testing.

The primary objectives of this research was to: 1) design an experiment to accurately measure the gas and water phase concentrations of ²²²Rn when the radon is in equilibrium between both phases; 2) minimize the error due to multiple analytical system calibrations by designing an experiment to utilize only a single system and thus remove a requirement for intercalibration; and 3) calculate the partition coefficient at different temperatures and salinities and compare them to values derived from the Weigel equation.

1.2.5 Collaborating Laboratory

This experiment was conducted in collaboration with Michael Schubert's research group at the Helmholtz Centre for Environmental Research (UFZ) in Leipzig, Germany. In order to ensure accuracy of the experimental data, both data sets were combined and the results were published in Environmental Sciences and Technologies (Schubert et al. 2012). The two laboratory approaches were based on different measurement methods and techniques producing independent data sets that were combined to minimize any experimental errors.

CHAPTER TWO

METHODS

2.1 Instrumentation

To re-evaluate the relationship $K_{w/air} = f(T, S)$, two independent experimental setups were designed and run by two individual laboratory groups. One experimental setup (as described below) was run at Florida State University, Department of Earth, Ocean and Atmospheric Sciences in Tallahassee, Florida, while the other experiment was conducted by our colleague, Michael Schubert, at UFZ, Helmhotz Centre for Environmental Research in Leipzig, Germany.

2.1.1 Detector

The detectors employed were RAD7 (Durridge Company, Inc., Billerica, MA) electronic radon detectors. The RAD7 utilizes a planar silicon detector that determines the ²²²Rn activity continuously by measurement of the electrostatically attracted ²¹⁸Po and ²¹⁴Po daughter isotopes (Xu et al. 2009). The RAD7 detector was run in 'sniff' mode with a sensitivity of 0.2 cpm/pCi/L (7.4 cpm/Bq/m³) and a dynamic range of 0.1 – 10,000 pCi/L (3.7- 370,000 Bq/m³). To remove a possible source of error, the same RAD7 was utilized during all analyses at FSU of both the gas phase and liquid phase. Thus, even if the RAD7 had a calibration error, the ratio of the radon-in-air to radon-in-water would be unaffected.

Durridge Company designed and manufactured multiple accessories with the primary purpose to measure radon in different mediums with maximum efficiency. Two accessories used in our work are the RAD-H₂O, a direct radon-in-water measurement tool and the RAD-AQUA, a liquid spray chamber used for continuous measurement. The RAD-H₂O utilizes a predetermined air and water volume and aerates the water sample to remove the radon with a predetermined and calibrated stripping efficiency. By utilizing this efficiency, the RAD7 can accurately measure water ²²²Rn activities from about 50 pCi/L (185 Bq/m³) and upwards of 10⁵ pCi/L (3,700,000 Bq/m³) in approximately 30 minutes. The RAD-AQUA circulates a closed gas loop inside an exchanger chamber with an aerosolized open loop water supply. The ²²²Rn concentration of the closed gas loop will reach equilibrium with the ²²²Rn concentration of the water, depending upon its partition coefficient which is a function of the temperature and salinity. An Onset Hobo data logger and temperature probe (Onset Computer Corporation, Cape Cod, MA) is inserted with the probe protruding into the aerosolized spray field to continuously measure the liquid temperature. For pure water, a manual calculation can be made to determine the liquid phase radon concentration by calculating the partitioning coefficient using the logged temperature and the Weigal equation and multiplying it by the radon-in-air concentration given directly by the output of the RAD7 detector.

2.1.2 Temperature Control

Due to the temperature dependency on the partition coefficient, a constant temperature was maintained for each experiment. For one set of experimental data points, a temperature of \sim 1.6 °C was achieved by submerging the container in an ice bath. It was observed that, while the ice bath was at 0 °C, the insulating properties of the sample container combined with the ambient room temperature resulted in an internal temperature of nearly 1.6 °C for at least 3 hours. For another set of experimental data points, a temperature of 27 °C was selected and was maintained above standard room temperature by employing a submersible heater and controller. A Won Brother D-58 (Won Brothers, Inc., Fredericksburg, VA) temperature controller and an 800-watt submersible titanium heater were used with a reported precision of ± 1 °C. A titanium heater

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was specifically chosen over a standard glass or ceramic heater for its ability to handle a thermal shock when placed directly in a 0 °C liquid solution and set to increase the temperature to 27 °C. An Aqueon 950 (Central Garden & Pet, Franklin, WI) circulation pump was utilized to ensure a constant mixing during the heating phase eliminating the possibility of any temperature gradient occurring in the sample container (Fig. 3).

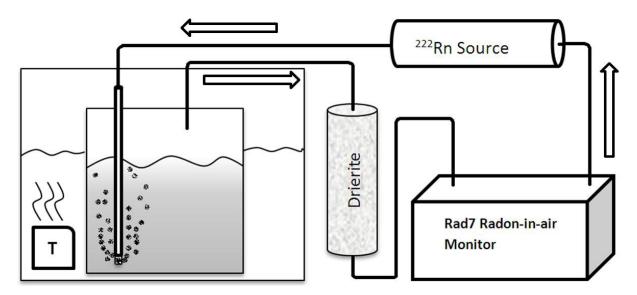


Figure 3. Direct Measurement (Volume Independent) Method Radon Introduction and Equilibrium. Radon is circulated from the generator into the bottom of the sealed container. From there, the radon is bubbled from the water phase into the gas phase and proceeds out the top of the container into the desiccant (Drierite) column. As the relative humidity of the air sample is decreased to <10%, the air enters the Rad7 Radon-in-air monitor for concentration measurements and the cycle repeats until equilibrium is established.

An Onset Hobo Data Logger H08-001-02 single-channel temperature logger with the TMC6-HC 6' stainless steel temperature probe was utilized to monitor temperature over the course of each experiment. Based upon instrument specifications, the TMC6-HC maintained a < 0.1° C drift per year with a typical response time of 15 seconds in disturbed water. The TMC6-HC specifications stated an accuracy of $\pm 0.5^{\circ}$ C between 0° C and 30° C and resolution of <0.5° C between 0° C and 40° C.

2.1.3 Radon Source

The original experimental design used local radon-rich tap water (~10,000 Bq/m³) as a radon source by spraying the water through a RAD-AQUA water exchange chamber. Utilizing a closed loop air supply and an open water supply, ²²²Rn gas reached equilibrium with the concentration of ²²²Rn in the liquid phase with respect to the partition coefficient (Burnett et al., 2001). Depending upon the water flow rate, water-air equilibrium normally take about 20-40 minutes. One problem with this approach was the somewhat low ²²²Rn concentration in relation to the amount that would later have to be analyzed in small (250 mL) water samples by the RAD-H2O. Because of that deficiency, this setup was replaced with a radon canister source containing ²²⁶Ra-charged impregnated manganese fibers with enough radium to produce a radon-in-air activity of ~ 100,000 Bq/m³ after an ingrowth time of a few days (Peterson et al., 2009).

The major benefit of using the radon canister generator was that it easily generated a higher activity of ²²²Rn than what was available in the local water supply. These higher activities increased the precision of the radon-in-water measurements made by the RAD-H₂O. Another major enhancement of the canister design over the exchanger was the equilibrium time when ²²²Rn was introduced into the system. The canister released the radon gas immediately and the equilibration time during experiments was about 20 min rather than the 40 min required for the tap water exchanger. The theoretical time for radioactive equilibrium between ²²²Rn and ²¹⁸Po is about 15 minutes, or roughly 5 half-lives of ²¹⁸Po (T_{1/2} = 3 min). One minor disadvantage of the canister source was time required to allow the ²²²Rn ingrowth from ²²⁶Ra decay after each experiment. During this time the exchanger system could constantly generate more ²²²Rn from a virtually never ending supply in the local tap water.

2.1.4 Salinity Adjustments

The seawater used during experimentation was synthetically manufactured by saturating radium-free deionized water with a synthetic salt called "Instant Ocean," which mimics the natural constituents of seawater. The major cations of "Instant Ocean" are 594 mmol kg⁻¹, while natural seawater major cations are 607 mmol kg⁻¹. The major anions of "Instant Ocean" totaled 569 mmol kg⁻¹, while the anions of natural seawater are 608 mmol kg⁻¹ (Atkinson 1997). Based upon the similarity in these values, "Instant Ocean" was utilized to produce the synthetic sea water we used for experimental purposes. This solution was then passed through a spun acrylic filter to remove any undissolved constituents to produce a fully saturated stock solution. Utilizing an YSI Model 63 (Xylem Inc., Yellow Springs, OH) probe, the conductivity, and corresponding salinity of the stock solution was determined. By diluting the stock solution with radium-free deionized water, it was possible to make synthetic seawater of various salinities which were again checked with the YSI probe.

2.1.5 Sample Containers

The sample container used was an adaptation of the design from Stringer and Burnett (2004), with the additional modification of an internal temperature probe. The design utilized a 6L high-density Nalgene container with a modified 3 port cap. The cap was connected via tubing to sized quick disconnects that allowed easy setup and dismantling. The different sized disconnects allowed the system to be setup quickly without the possibility of attaching the input and output tubes to the wrong sides. The underside of the cap was designed so that one port was plugged because it was not used, one port was left exposed to the gas space at the top of the container, and the other was attached to a length of tubing and finished off with a sparging stone

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extending by tubing to the bottom of the container. A hole was drilled into the flat top of the container to allow a cable gland to be screwed in and then sealed with a silicon epoxy. The addition of the cable gland allowed the temperature probe to be inserted into the bottle and then formed an airtight seal around the cable, allowing a constant temperature reading of the water inside the container without exposing the system.

2.2 Experimental Approach

2.2.1 Volume Independent (Direct Measurement) Method

The initial setup utilized the RAD7 detector to measure the gas phase concentration of the system, while a radon extraction line was utilized to measure the radon dissolved in the liquid phase. This design was determined to be undesirable due to multiple factors including: an intercalibration required between the two systems (RAD7 and Rn line/scintillation cell); a high precision measurement of both the liquid and gas volumes; a long sampling time which required 90 minutes of sparging the sample on the radon line, a 3 hour ingrowth hold time before counting and a 60 minute count time; along with the task of ensuring no gas escaped during transfer from one system to another.

This setup was replaced with the RAD-H₂O radon-in-water setup, which had a radon-inwater response time of 10 minutes followed by four 5 minute counts, excluding the system purge time to minimize background. Utilizing a stacked Drierite and activate charcoal filter, background count rates on the RAD7 were lowered to 2 ± 2 cpm in about 40 minutes, depending upon the activity of ²²²Rn in the system prior to the purge. When the container was connected in the initial sparge configuration, the RAD7 was connected in series with the radon generator, sample container and a Drierite drying tube. Air was pumped out of the RAD7, via the internal

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air pump, and entered the radon generator canister where it then mixed with the ²²²Rn enriched air and was transferred out of the generator and into the sampler container inlet (Fig. 3). From there, the ²²²Rn travels down the tubing, through the cap and into the tubing attached to the sparging stone. It then continued bubbling into the liquid phase, and exited the liquid into the gas phase and continued back through the drying tube and again into the RAD7. At this point, the ²²²Rn concentration decreased as some of the radon in the gas dissolves into the sample water. When equilibrium in the system was reached (the observed count rate leveled off), seven 5 minute counts were conducted of the gas concentration and recorded. The middle count time was set as the sample time and the values were averaged. The connection was then placed in a reverse configuration using a different set of tubing specially designed to pressurize the headspace with high purity helium gas, which in turn forced the bottom water through the sparging stone and into a special connection that led into a 250 mL collection bottles that are used for the RAD-H₂O analysis (Fig. 4). This efficiently allowed the collection of ²²²Rn rich sample water for analysis without risk of exposing the sample to the environment and losing ²²²Rn.

2.2.2 Analysis of the Water Radon Concentration via the RAD-H₂O

Water samples in specially designed 250-mL bottles are then tested on the RAD-H₂O radon-in-water setup. After the first 4 sets of 5-minute counts are completed, an additional cycle was conducted and the 8 results were averaged. Samples were collected in triplicate and analyzed. The 24 resulting counts from all 8 sets of 5 minute measurements were analyzed and averaged. All values were then decay corrected back to the gas concentration sampling time.

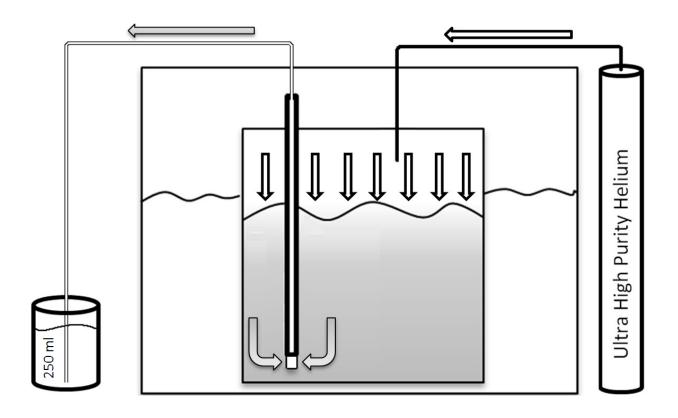


Figure 4. Direct Measurement (Volume Independent) Method Water Sample Extraction. The headspace of the container is pressurized with inert helium, extracting water at the bottom of the sample container into a 250 mL vial for radon concentration measurements on the RAD-H₂O.

2.2.3 Volume Dependent (Indirect Measurement) Method

This experimental setup was used in an attempt to determine the partitioning coefficient by measuring the changes in the ²²²Rn gas concentration by altering only the temperature in a closed system. The challenge of this experimental setup was the necessity to calculate precise volumes for each part of the system. Physical measurements were conducted to measure the entire volume of the sample container, then subtracting out water volume (*Vw*) to calculate the volume of gas in the sample container (*Vg2*). Calculating the volume of gas in the system minus *Vg2* (*Vg1*) was done by measuring 2 equilibrium points and utilizing the equation,

$$Vg1 = \frac{(Cw1*Vw) + (Cequ2*Vg2)}{Cequ1 - Cequ2}$$
(Equation 4)

Cequ1 was calculated by running the experiment in a closed gas loop, where the volume of gas was *Vg1*. *Cequ2* was calculated as the gas concentration by running the experiment after introducing the sample container, where the volume of gas is the sum of *Vg1* and *Vg2*. The gas was initially run through the generator, into the Drierite tube and into the RAD7, then out of the RAD7 and back into the generator. At the start of the experiment, T(0), the generator was bypassed and all the ²²²Rn at this point was unsupported (Fig. 5). This experiment relied on the change in the total concentration of ²²²Rn, therefore, it is important that all ²²²Rn in the system be unsupported so any decay loss could be accounted for. After the ²²²Rn value stabilized, the loop was opened up to the sample container vessel. The ²²²Rn concentration would suddenly drop as it partially dissolves into the liquid and is diluted by the gas headspace of the container. From there, it continued through the Drierite tube and into the RAD7 where it was analyzed and the cycle was continuously repeated.

In one experiment a sample container (23L Nalgene Carboy) with a modified 3 port cap setup was used in the same manner as before. A small portion of volume was unaccounted for in Vg1 due to the tubing bypass loop, however, this volume was negligible compared to the entire gas volume in the system because of minimal length and the diameter of the tubing. Cw1 was calculated by running the setup with fresh deionized, radium free water and utilizing the Weigel equation to determine the water concentration by using the gas concentration of the entire setup (Cequ2) and temperature. After the gas concentration and temperature reach equilibrium, a number of sample data points were collected and the temperature was then changed. Once the gas concentration and temperature reached a new equilibrium, a number of sample data points were collected and the new water concentration was labeled Cw2, while the new gas concentration was labeled Cequ3. By replacing Cw1 with Cw2 and Cequ2 with Cequ3, Vg1

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could be calculated using equation 5. By mass balance, we should be able to calculate Cw1 and Cequ2 or Cw2 and Cequ3. Once Vg1 had been accurately measured, the water concentration could be calculated utilizing the equation,

$$Cw1 = \frac{Vg1(Cequ1 - Cequ2) - (Cequ2 * Vg2)}{Vw}$$
(Equation 5)

By modifying the temperature and establishing a new equilibrium point, the new equilibrium gas concentration can be used to calculate the water concentration. In principle, by running the experiment at set salinity values, the partitioning coefficient could be calculated throughout any temperature range that the experimental setup allowed.

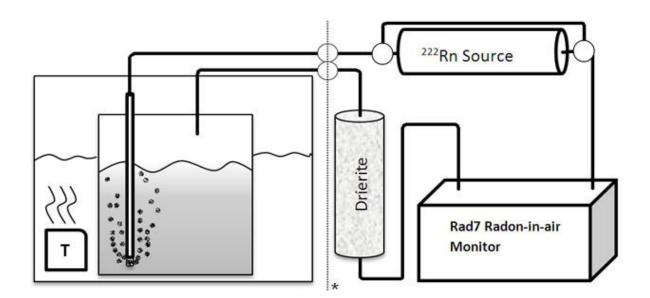


Figure 5. Volume Dependent (Indirect Measurement) Method Experimental Setup. The complete system includes both the first half of the experiment and the second (denoted at the line marked *). For the first half of the experiment (everything left of the * line including a bypass between the two circle to complete the loop), the radon canister is opened and radon is introduced into the RAD7 detector. After 10 minutes, the radon canister is bypassed (utilizing the tubing above the canister) and the concentration of radon in the system is measured at Cequ1 with a volume of Vg1. The second half of the experiment involved removing the bypass, at the line denoted as *, and introduce the sample container to the cycle. Radon from the system is introduced to the bottom of the water sample and bubbled into a measured volume (Vg2) of gas in the headspace. The gas is passed thru the desiccant column until the relative humidity is <10% and measured via the Rad7 detector. Once equilibrium is established and multiple data points are collected, the temperature of the sample container is changed and the process continues until equilibrium is established again.

2.2.4 Setup at Helmholtz Centre for Environmental Research (UFZ) Method (Schubert et al. 2012)

The experimental setup at UFZ in Leipzig, Germany differed from the Florida State University (FSU) setup, by utilizing a cluster of stripping units and a radon-in-air monitor to sparge 4 independent water samples of different salinities with the same radon-rich air. The radon-in-air monitor (also a RAD7 detector) recorded the radon concentration of the gas phase once equilibrium was reached. The UFZ experiment also used NaCl salt instead of a seawater substitute salt (like Instant Ocean) used in the FSU experiment. Unlike the FSU experiment, which utilized a water bath to moderate the temperature of the container vessel, the UFZ experiment was conducted inside a laboratory refrigerator and inside a hot-air cabinet to control the temperature of the entire apparatus. Each radon stripping unit was 150 mL and filled with a water of a known salinity without any headspace. To measure the dissolved radon concentration, the glass stripping units were analyzed with an ORTEC-Gamma-X HPGe coaxial gamma detector. The activity was measured by γ -spectrometry, which was then divided by the water volume to determine the concentration of radon dissolved in the water phase.

CHAPTER THREE RESULTS AND DISCUSSIONS

3.1 Error Minimization

3.1.1 Radium 226 in Drierite

Due to the requirement to minimize humidity in the RAD7 detector, Drierite was used as the standard desiccant material. Drierite is a desiccant compound of anhydrous CaSO₄, manufactured from gypsum along with a color indicator to visually indicate when the material is saturated and needs to be replaced. In order to determine the ²²²Rn partitioning coefficient, all methods of ²²²Rn production had to be monitored. Due to the possibility of radium present in the Drierite (Guimond and Hardin 1989), a RAD7 was placed in a closed loop configuration with a standard Drierite column and left to run for 1 week (Fig. 6). The rate of ²²²Rn release due to ²²⁶Ra decay, in a normal amount of Drierite, was observed to be 7x10⁻⁴ Bq/m³/min. During experimental testing with a maximum test time of 320 minutes and average total radon concentration of 90,000 Bq/m³, the ²²²Rn due to ²²⁶Ra decay in Drierite was calculated to be about 224x10⁻³ Bq/m³, or roughly 2.5x10⁻⁴% of the total radon in the system. This value is considerably smaller than the analytical error of the setup and any radon production due to Drierite was determined to be negligible.

3.1.2 Radium 226 in Instant Ocean

Another possible source of unwanted radon production would be from radium present in Instant Ocean. A 100 cc can was filled with 135.29 grams of Instant Ocean and placed on a coaxial gamma detector and analyzed for 122 hours. The 295.0, 351.9 and 609.3 keV photopeaks were then monitored for ²¹⁴Pb and ²¹⁴Bi. Based upon the total counts per channel, the activity of ²¹⁴Pb and ²¹⁴Bi was determined and the ²²⁶Ra activity was calculated to be 0.09 Bq/kg. Based on this analysis of Instant Ocean, the ²²²Rn in-growth due to ²²⁶Ra was also determined to be negligible.

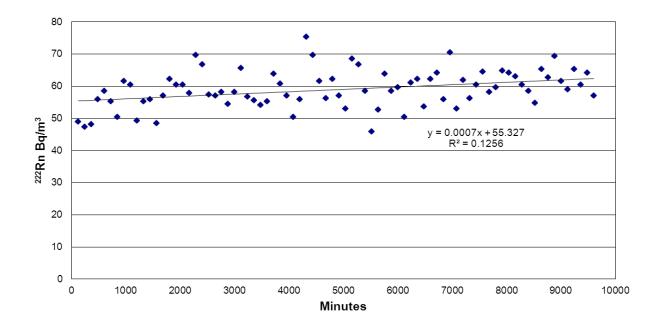


Figure 6. Radon Ingrowth in Drierite. Due to the possibility of radium in the calcium sulfate desiccant material, a canister was sealed and placed in line with a RAD7 detector. The radon concentration was measured for 10,000 minutes (roughly 7 days) and the radon ingrowth rate was determined by the slope of the trend line.

3.1.3 Calibration of the RAD-H₂O system

To increase the sensitivity of the RAD7 detector, a unit with an oversized dome was used but not calibrated for the RAD-H2O by the manufacturer. Standard RAD7s, with a normal sized dome, do not require any additional calibration for the RAD H2O measurements, i.e., the calibration is set by the manufacturer. An independent calibration correction factor was determined by running the RAD AQUA exchanger off a laboratory faucet and using the ²²²Rn gas value and the Weigel equation to determine the ²²²Rn water concentration. RAD-H₂O water samples were collected in triplicate from the tap water and run through two complete cycles on the RAD-H₂O setup. The average RAD AQUA value were then divided by the average RAD-H₂O value to create a correction factor used to correct all RAD-H₂O measurements. This experiment was repeated 5 times and those values averaged to product the calibration factor of 1.30 ± 0.14 (Fig. 7). All RAD-H₂O samples were run on the same RAD7 detector, and multiplied by this calibration factor to correct for the internal calibration error.

3.2 Volume Independent (Direct Measurement) Method

3.2.1 Experimental Results

Utilizing the methodology outlined in Chapter 2.2.1, 7 experimental data points were collected at a mean temperature of 26.7 °C and 9 were collected at 1.6 °C using synthetic salt water. Salinity values varied between 0 - 44.4 ppt using Instant Ocean as a natural seawater substitute. An additional data point was collected at 1.6 °C using natural salt water collected from Little Lagoon in Gulf Shores, Alabama (Fig. 8).

When the partitioning coefficient was plotted versus salinity, a linear trend line resulted represented the salinity effect at each temperature. Comparing the slopes of the linear trend lines, it becomes clear the salinity effect is more pronounced in colder waters. This can explain why the effect was not noticeable in previous comparisons, due to the majority of the published tests being done in warm, moderately low saline waters from the Gulf of Mexico and other tropical areas. The single data point from a non-synthetic salt water sample corresponds well with the experimental results and shows that there is no experimental bias due to the synthetic salt being used.

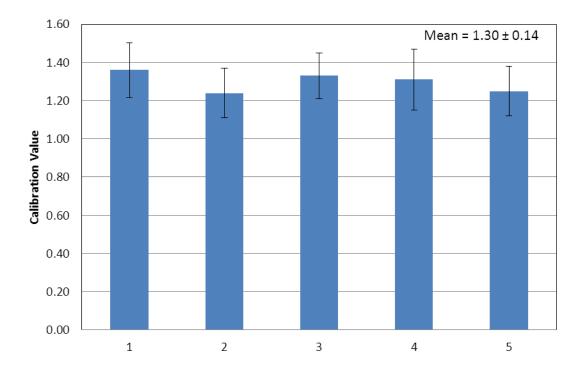


Figure 7. RAD H₂O Calibration. For an increased resolution, the detector dome inside the RAD7 was replaced with a larger volume dome. Since the RAD-H₂O relies on the air volume inside the detector, a calibration correction factor was calculated by dividing the RAD-AQUA measurements of standard radon rich laboratory water with the concentration measurement of the RAD-H₂O for 250 mL of the same water. All RAD-H₂O measurements were multiplied by this calibration factor.

3.2.2 Published Results

These experimental results were combined with the experimental data conducted in Germany and published in Schubert et al (2012) (Fig. 9). Figure 9A combines all data points, including salinity values used by UFZ, while figure 9B included specific data points limited to natural salinity values (0-50‰) seen in most natural waters. The published parameters for equation 2 are listed in Table 1 and were calculated using multiple linear regression analysis (Schubert et al. 2012). By utilizing equation 2 and the parameters in Table 1, a 3 dimensional representation of the salinity and temperature dependency for the radon partitioning coefficient was mapped out (Fig. 10). A 1:1 comparison of the Schubert et al. method (utilizing table 1 parameters in eq. 2 and 3) and the Weigel equation (eq. 1) for pure water (salinity = 0) shows

excellent agreement between the two in a temperature range of 0-27 °C (Fig. 11). The model approach produced values which may slightly underestimate the partitioning coefficient at colder temperatures and overestimate them at warmer temperatures, but the difference between the Schubert et al. method and the Weigel equation would be overshadowed by any salinity effects, especially in cold, high salinity water bodies.

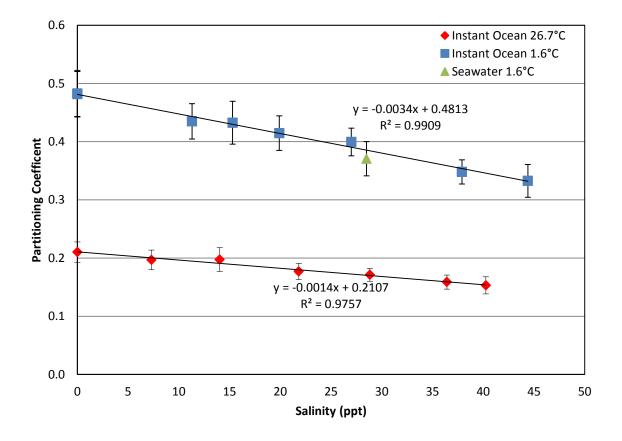


Figure 8. Experimental Results. The calculated partitioning coefficient at 2 temperatures (26.7 °C and 1.6 °C) of 14 synthetic seawater and 1 natural seawater samples vs. salinity.

| Table 1. Radon parameters for the Weiss Equation. The calculated a ₁ to b ₃ parameters for the Weiss equation |
|--|
| (Eq. 2) to determine the Bunsen coefficient in a salinity range of 0-54‰ and 0-360‰ and a temperature range of |
| 273-323 K (0 to 50 °C) (Schubert et al. 2012). |

| T [K] range | S range | a_1 | a ₂ | a ₃ | b_1 | b_2 | b ₃ |
|-------------|---------|--------|----------------|----------------|---------|--------|-----------------------|
| 273-300 | 0-54 | -77.91 | 122.82 | 32.13 | -0.4656 | 0.3049 | -0.0504 |
| 273-323 | 0-360 | -76.14 | 120.36 | 31.26 | -0.2631 | 0.1673 | -0.0270 |

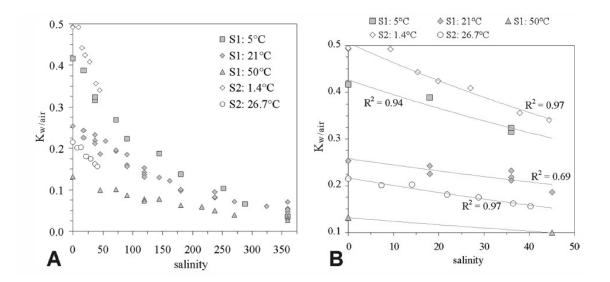


Figure 9. (A) UFZ and FSU Combined Results (Entire Salinity Scale). All experimental data from both the UFZ and FSU experiments. FSU experimentation was limited to 45 ppt salinity while the UFZ experimentation was extended out to 350 ppt., (B) UFZ and FSU Combined Results (Restricted Salinity Scale). Experimental data restricted to 0-45 ppt salinity. The experimental error of determining K_{w/air} was estimated to be 7.8%, based on error propagation (Schubert et al. 2012). Reprinted with permission from (Schubert et al. 2012). Copyright 2012 American Chemical Society.

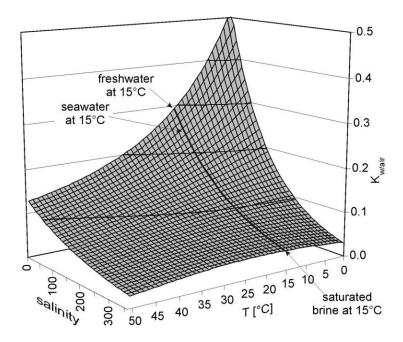


Figure 10. Weiss Equation with Radon Parameters (Schubert et al. Model). Utilizing the a₁ to b₃ parameters in Table 1 and the Weiss Equation (Eq. 2 and 3), a 3-dimension representation of the temperature and salinity effect on the partitioning coefficient was generated. As hypothesized, the largest change in the partitioning coefficient (as a function of salinity) is at lower temperatures (Schubert et al. 2012).

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3.3 Volume Dependent (Indirect Measurement) Method

The volume dependent method was designed as an attempt to rapidly collect temperature data points at a single salinity instead of re-setting the experiment each time. By maintaining a closed system and analyzing the change in the ²²²Rn gas concentration as the temperature changes, it theoretically should be possible to calculate the ²²²Rn liquid concentration by mass balance and determine the partitioning coefficient at each temperature. Once the gas concentration was calculated, the sample container was introduced to the system, where a dilution with radon free air and water caused the concentration to decrease greatly. After this primary dilution occurred, the radon concentration further decreased due to radon gas dissolving into the liquid medium. Once equilibrium was established, any changes in the gas concentration would be a direct result of the temperature dependency of the partitioning coefficient. Figure 12 portrays the results from one run. The radon concentrations are shown at the original gas concentration (Equ1), the equilibrium concentration after dilution and partitioning with the water inside the container (Equ2) and the equilibrium concentration after a temperature change (Equ3).

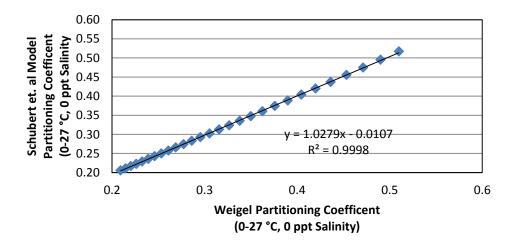


Figure 11. Weigel vs Schubert et al. Method. A 1:1 ratio comparison of the Schubert et al. Model Partitioning coefficient, calculated from the Table 1 a_1 to a_3 parameters (b_1 to b_3 are only required for salinity >0 ‰) and the Weiss equation (Eq. 2 and 3) vs the Weigel equation (Eq. 1) from 0-27 °C.

The main problem, as described in chapter 2.2.3, was accurately measuring the volume, known as Vg1. The experiment was conducted twice at 0 ppt salinity and temperatures between 2° C and 30° C, in an attempt to calculate the Vg1 by utilizing Equation 5. Due to the nature of the experimental setup, high precision volume measurements had to be taken for Vg2 and Vw, and any minor errors on these volume measurements had a large impact on the results. Figure 13 depicts the plotted data points from two runs, and shows some disagreement between the experimental values and the Weigel equation (deviation from the 1:1 ratio line). Due to this disagreement, different Vg1 values were being calculated at each temperature point which made it impossible to get an accurate Cw measurement. Several factors could have led to a failure in this experimental approach including: incorrect temperature values recorded by the temperature probe, not precise volume measurements and any possible escape of ²²²Rn gas from the system.

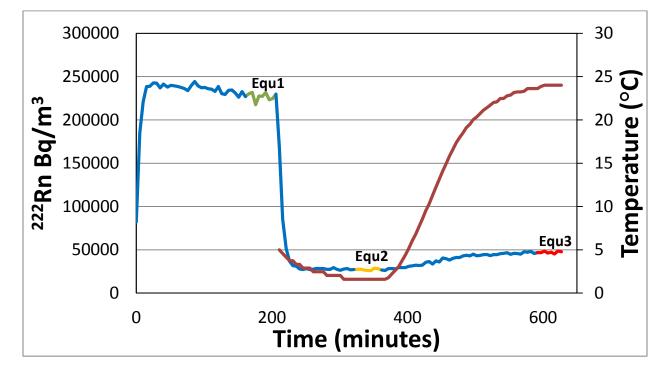


Figure 12. Volume Dependent Experimental Run. The plotted radon concentration of the gas phase, during a volume dependent (indirect measurement) experimental run, as a function of temperature. Equ1 corresponds with the gas phase concentration of radon gas during the first half of the experimental (where volume is expressed as Vg1). The major drop off in radon concentration after Equ1 is due to the introduction of radon free gas (Vg2) in the container vessel. After a certain amount of time, additional radon is dissolved into the water phase and equilibrium is established (Equ2). The red line plots the internal temperature of the water inside the sealed container. To get a second set of experimental data points, the internal temperature of the container is increased to a new point and radon equilibrium between the gas and water phase is reestablished (Equ3).

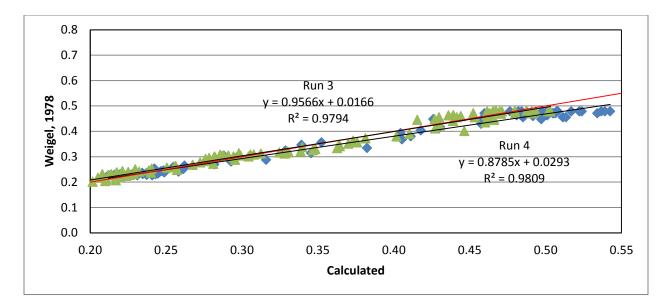


Figure 13. Volume Dependent Vs. Weigel Equation. Runs 3(green) and 4(blue) were the most stable out of 5 experimental runs, so those were plotted against the Weigel partitioning coefficient. The general disagreement between the experimental results and the Weigel calculated partitioning coefficient (when compared to a 1:1 line depicted in red) emphasized the inability to calculate an accurate and reproducible Vg1 value (Eq. 4).

CHAPTER 4

CONCLUSIONS

The main objective of this experimental work were to determine the extent of the salinity effect on the partitioning coefficient for radon gas. This work has showed that the salinity effect, when compared to the historical Weigel equation, is greater than previous assumed and that a partitioning coefficient that included salinity effects was necessary to properly assess radon concentrations in marine waters.

An experimental setup was designed to map out the salinity effect at different temperatures and we combined our results with those of M. Schubert to obtain a relationship of the partition coefficient as a function of salinity as well as temperature.

4.1 Volume Independent (Direct Measurement) Method

The experimental setup confirmed there is a salinity effect greater than previously assumed and produced a model that can be applied to direct measurements taken in the field (Equation 3). An alternate direct liquid measurement setup was conducted in Leipzig, Germany using γ -spectroscopy in sealed liquid vials (Schubert et al 2012) and the results verified and validated my results using the RAD-H₂O.

Based on the final results (depicted in figure 10), it is easy to identify why salinity was not previously thought to play a crucial role in the partitioning coefficient for marine waters. The difference in the partitioning coefficient is not as pronounced in warm temperatures as it is in colder temperatures. Since the original experimentation was done in the warm subtropical to tropical waters of the Gulf of Mexico and elsewhere, the experimental error of the radon measurements (often $\sim 10\%$) concealed any error in the partitioning coefficient calculated using the Weigal equation designed for pure water.

4.2 Volume Dependent (Indirect Measurement) Method

The experimental setup for the volume dependent method never gave reproducible results. The mathematical theory (eq. 4 and 5) appears sound, but multiple factors led to an inability to measure an accurate Vg1 volume. Communication with the manufacturer of the RAD7 radon-in-air monitor showed that there is a known small leak in the RAD7 air pump. Usually this leak is so insignificant that any error due to it is negligible, but this is not true when attempting to calculate precise volumes. Another factor that led to the inability to measure the volume was the non-uniformity of Drierite desiccant material. Without a precise uniformity and measurement of the weight of the desiccant material, the volume of the column would change each time the material was replaced. Originally this was thought to be negligible, but upon experimentation, it was determined that any change in the Vg1 volume (even very small) would lead to large changes in the calculated concentration of radon in water.

APPENDIX A

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BIOGRAPHICAL SKETCH

Eric B. Lieberman was born in 1985 in Coral Springs, Florida. He grew up and pursued schooling in Parkland, Florida. With an interest in Chemistry and Physics, he took advanced placement (AP) courses in Chemistry and Physics in high school at Marjory Stoneman Douglas High School in Parkland, Florida. He continued his studies at Florida State University in Tallahassee, Florida eventually completing his B.S. degree in Chemical Science with a minor in Physics. During this time, he worked as an OPS technician in the Academic Diving Program at Florida State University, conducting support for research missions and offering technical expertise in equipment utilization. Upon graduation, he was employed by the Florida Department of Health as an Environmental Specialist in the Bureau of Radiation Control. His work experience with the bureau led to a strong interest in the way radioactive materials physically and chemically bond and eventually that interest led to research.