# **BIG BOTTLE SYSTEM**

High Sensitivity Radon in Water Accessory for the RAD7 With Aerator Cap Revision B

**User Manual** 





## INTRODUCTION

The Big Bottle System is an accessory to the RAD7 radon monitor that enables you to measure radon in water over a concentration range of from 1 pCi/L to greater than 10,000 pCi/L. For higher ranges, the standard RAD  $\rm H_2O$  accessory is recommended.

The complete system, including the RAD7, is portable and battery operated, and the measurement is moderately fast. Even for the lowest radon concentrations you can have an accurate reading of radon in the water within two hours of taking the sample. The Big Bottle System gives results after 120 minutes analysis, or less, with a sensitivity that far exceeds that of liquid scintillation methods. The method is simple and straightforward, though more complicated than with the standard RAD  $H_2O$ . There are no harmful chemicals to use. Once the procedure becomes familiar and well understood it will produce accurate results with minimal effort.

It is assumed that the user has a good, working knowledge of the RAD7. If both the RAD7 and the Big Bottle System are new to the user, then time should be spent learning how to make good measurements of radon in air with the RAD7 before embarking on radon in water measurements. Instructions for RAD7 operation specifically with the Big Bottle System are given in this manual but, for more detail about the instrument and its use to measure radon and thoron in air, the reader is referred to the RAD7 manual.

Introduction 2

## TABLE OF CONTENTS

INTRODUCTION					
TABLE OF CONTENTS					
1 GETTING STARTED		5			
1.1 Unpacking		5			
1.2 General Safety Instructions		5			
1.3 Taking a Look		5			
Fig. 1 The Big Bottle System Glass Jug	Kit (left) and Soda Bottle Kit (right)	5			
Fig. 2 Temperature Logger Kit (left) and	d the Retort Stand Kit (right)	6			
Fig. 3 Big Bottle System Schematic		7			
1.4 Running a Test		8			
1.4.1 Preparing the RAD7		8			
1.4.2 Collecting a Sample		8			
1.4.3 Setting up the equipment		8			
Fig. 4 Aerator and Bottle assembly wit	n Temperature Logger	9			
1.4.4 Running the test with desiccant		9			
Fig. 5 Aeration in progress		10			
1.4.5 Running the test with the DRYSTIK		10			
Fig. 6 Big Bottle System with DRYSTIK		10			
1.4.6 Finishing the Test		11			
1.4.7 Interpreting the results Fig. 7 Big Bottle System printout		11 12			
rig. 7 big bottle system printout		12			
2 BIG BOTTLE SYSTEM MEASUREMENT	TTECHNIQUE	13			
2.1 The Closed Loop Concept		13			
2.2 Desiccant		13			
2.3 Purging the System		13			
2.4 Background and Residuals		14			
3 CALCULATING RESULTS		16			
3.1 How Calculation Is Made		16			
3.2 Detailed Calculation		16			
3.2.1 Definitions		16			
3.2.2 Known Values		16			
3.2.3 Head Space Loss		17			
3.2.4 Air-equivalent of water		17			
3.2.5 Calculation		17			
3.2.6 Correction for ambient/purge radon		17			

Table of Contents 3

	3.2.7 Radon Graph Interpretation	17
	3.2.8 Decay Correction	17
	3.3 Automatic Calculation with CAPTURE Software	18
	Fig. 8 The CAPTURE Big Bottle System configuration interface	19
	Fig. 9 Decay Correction Factors Table	20
4	ACCURACY AND QUALITY CONTROL	21
	4.1 Calibration of System	21
	4.2 Accuracy and Precision	21
	4.2.1 Sampling Technique	21
	4.2.2 Sample Concentration	21
	4.2.3 Sample Size	21
	4.2.4 Counting Time	21
	4.2.5 Temperature	22
	4.2.6 Relative Humidity	22
	4.2.7 Background Effects	22
	4.3 Comparison of RAD H2O and Big Bottle System with Other Methods	22
	4.4 Quality Assurance	22
	Fig. 10 Method Comparison	23
5	CARE, MAINTENANCE and TROUBLESHOOTING	24
	5.1 Warning on Pump Direction	24
	5.2 Warning on Tipping the Aeration Unit	24
	5.3 Stones Maintenance	
	5.5 Stories Maintenance	24
	5.4 High Humidity	24 24
	5.4 High Humidity	24
	5.4 High Humidity 5.5 Foaming	24 25
	<ul><li>5.4 High Humidity</li><li>5.5 Foaming</li><li>5.6 Plastic Bottle Denting</li></ul>	24 25 25
6	<ul><li>5.4 High Humidity</li><li>5.5 Foaming</li><li>5.6 Plastic Bottle Denting</li><li>5.7 Glass Jug Care</li></ul>	24 25 25 25
6	<ul><li>5.4 High Humidity</li><li>5.5 Foaming</li><li>5.6 Plastic Bottle Denting</li><li>5.7 Glass Jug Care</li><li>5.8 Technical Support</li></ul>	24 25 25 25 25
6	<ul> <li>5.4 High Humidity</li> <li>5.5 Foaming</li> <li>5.6 Plastic Bottle Denting</li> <li>5.7 Glass Jug Care</li> <li>5.8 Technical Support</li> </ul> BACKGROUND TO RADON-IN-WATER MEASUREMENT	24 25 25 25 25 25
6	<ul> <li>5.4 High Humidity</li> <li>5.5 Foaming</li> <li>5.6 Plastic Bottle Denting</li> <li>5.7 Glass Jug Care</li> <li>5.8 Technical Support</li> </ul> BACKGROUND TO RADON-IN-WATER MEASUREMENT <ul> <li>6.1 About Radon-in-Water</li> </ul>	24 25 25 25 25 26
6	<ul> <li>5.4 High Humidity</li> <li>5.5 Foaming</li> <li>5.6 Plastic Bottle Denting</li> <li>5.7 Glass Jug Care</li> <li>5.8 Technical Support</li> </ul> BACKGROUND TO RADON-IN-WATER MEASUREMENT <ul> <li>6.1 About Radon-in-Water</li> <li>6.2 Health Risks Due to Waterborne Radon</li> </ul>	24 25 25 25 25 26 26
6	<ul> <li>5.4 High Humidity</li> <li>5.5 Foaming</li> <li>5.6 Plastic Bottle Denting</li> <li>5.7 Glass Jug Care</li> <li>5.8 Technical Support</li> </ul> BACKGROUND TO RADON-IN-WATER MEASUREMENT <ul> <li>6.1 About Radon-in-Water</li> <li>6.2 Health Risks Due to Waterborne Radon</li> <li>6.3 Physical Properties of Waterborne Radon</li> </ul>	24 25 25 25 25 26 26 26 27
6	<ul> <li>5.4 High Humidity</li> <li>5.5 Foaming</li> <li>5.6 Plastic Bottle Denting</li> <li>5.7 Glass Jug Care</li> <li>5.8 Technical Support</li> </ul> BACKGROUND TO RADON-IN-WATER MEASUREMENT <ul> <li>6.1 About Radon-in-Water</li> <li>6.2 Health Risks Due to Waterborne Radon</li> <li>6.3 Physical Properties of Waterborne Radon</li> <li>6.4 Radon as a Tracer for Groundwater movement</li> </ul>	24 25 25 25 25 26 26 27 27

Table of Contents 4

## 1.1 Unpacking

First make sure the package contains everything you are supposed to have. Examine the case contents and verify that you have all the items shown in the packing list. If anything is missing, please call DURRIDGE immediately at (978) 667-9556 or email sales@durridge.com.

## 1.2 General Safety Instructions

There is nothing hazardous to the user in the Big Bottle System, but care should be taken to make sure that water never enters the RAD7. The check valve attached to the aerator should never be removed, as it protects the RAD7 in the event that the tube connections to the instrument are reversed.

## 1.3 Taking a Look

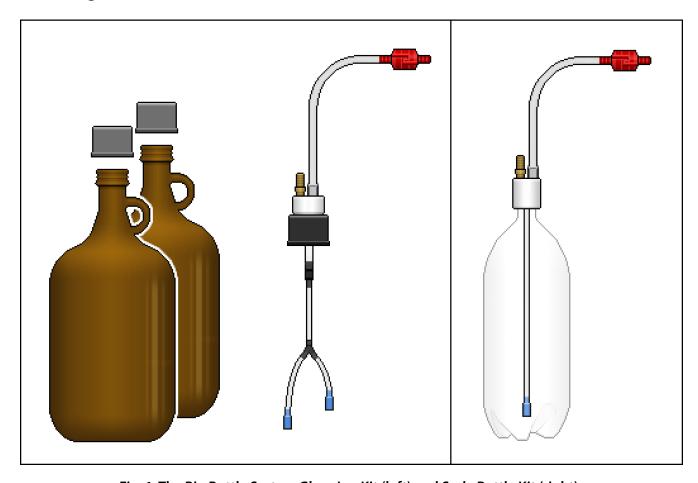


Fig. 1 The Big Bottle System Glass Jug Kit (left) and Soda Bottle Kit (right)

(Note: the plastic soda bottle is not included.)

The Big Bottle System consists of several components. Depending on whether the system is purchased for use with samples collected in a glass jug or a soda bottle, it will come with either the Glass Jug Kit (see Fig. 1, left) or the Soda Bottle Kit (Fig. 1, right.)

The Glass Jug Kit comes with two 2.5 L bottles and an aeration system, while the Soda Bottle Kit comes with only an aeration system. It is compatible with most 500mL - 2L pressurized PET soda bottles, which are sold separately.

If the Soda Bottle Kit is being used, be aware that large soda bottles with insufficient wall strength may become dented inward during aeration. Any indentation will reduce the volume of the bottle and displace water upward towards the desiccant. See Section 5.6 for further details and tips on how to avoid this problem.

Each aeration system includes a check valve and one or two aeration stones. Both the Glass Jug and the Soda Bottle systems include the Temperature Logger Kit and the Retort Stand Kit, which are detailed below. The Temperature Logger Kit (see Fig. 2, left

section) consists of a Lascar Temperature probe with a USB cable and necessary software, a hermetically sealed thermocouple, and an elastic, padded strap for attaching the thermocouple to the bottle. The Retort Stand Kit (see Fig. 2, right section) consists of a short stand, clamp, bubble trap, and tubing set with inlet filter.

Any Big Bottle system will require a RAD7 radon monitor, plus either a Laboratory Dryer or DRYSTIK. These items must be purchased separately.

An example of a fully assembled Big Bottle System is shown in Fig. 3 on the following page.

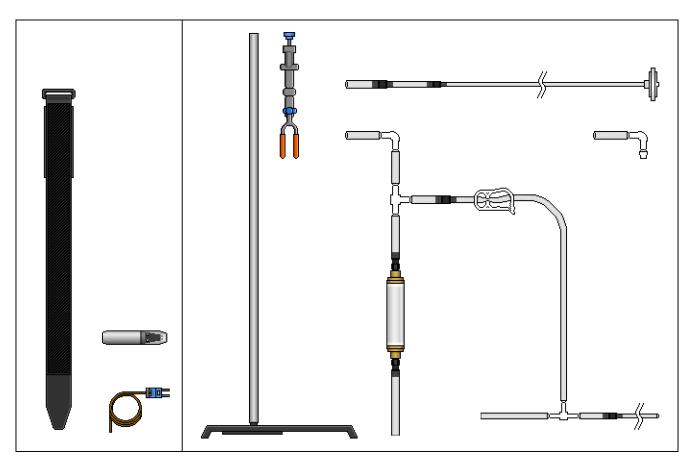


Fig. 2 Temperature Logger Kit (left) and the Retort Stand Kit (right)

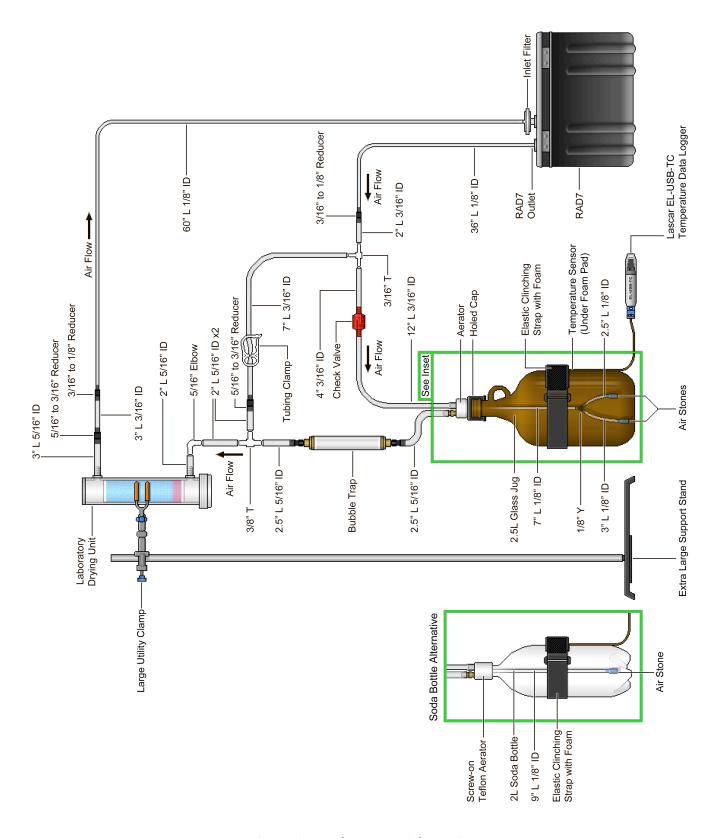


Fig. 3 Big Bottle System Schematic

## 1.4 Running a Test

#### 1.4.1 Preparing the RAD7

Before making a measurement, the RAD7 must be free of radon and dry. To achieve this, it should be purged for some time. A number of drying options may be chosen for the measurement itself, but it is convenient to use the large laboratory drying unit during the purging process, to save the small drying tubes for use with the actual measurement.

Hook up the laboratory drying unit to the RAD7 inlet, with the inlet filter in place (see RAD7 manual). Any tube connected to the outlet must be open to the air at the other end. Purge the unit with fresh dry air for ten minutes.

After 10 minutes of purging with dry air, push the [MENU] button, then push [ENTER] twice, to go to the status window, and push the right arrow button twice to see the relative humidity. If it is not yet down close to 10%, start purging some more. To conserve desiccant, after the first ten minutes or so of purging, you may connect the RAD7 outlet to the inlet of the laboratory drying unit, thus forming a closed loop. This will continue to dry out the RAD7 but will no longer purge the unit with fresh air.

If the RAD7 has not been used for some time, or if it has been left without the small tubing bridge in place between the air inlet and outlet, then it may take an hour or more to dry it out. Once it has thoroughly dried out, however, just 15 minutes of purging and drying between measurements of different samples will generally be sufficient.

#### 1.4.2 Collecting a Sample

Getting a good sample requires care and practice. Sampling technique, or lack of it, is generally the major source of error in measuring the radon content of water. The water sampled must be a) representative of the water being tested, and b) delivered to the Big Bottle without ever being in contact with air.

To satisfy (a), make sure that the sample has not been through a charcoal filter, or been sitting for days in a hot water tank. To test a well, choose a faucet at the well, or outside the house, before the water enters any treatment process. Run the water for several minutes, to make sure that the sample comes fresh from deep in the well.

To satisfy (b), one method is to attach a tube to a faucet, and insert the tube into the bottle so that it reaches the bottom. Fill the bottle and allow water to overflow for a while, to ensure that it is filled with water that has not lost any radon to the surrounding air. After filling a 2.5L glass jug, immediately decant a few mL so that the water level is down near the flange below the thread on the neck of the bottle. Then replace the cap. The air-space in the bottle provides an expansion buffer and prevents the bottle from being broken by expansion of the water as it warms up to room temperature.

Another method, suitable for sampling in open water, is to lower the empty bottle upside down to the depth required and then inverting the bottle so that the air inside is displaced by water. This method has a disadvantage in that the air in the bottle, when it leaves, will take some of the radon out of the water as it replaces the air in the bottle, so that the sample may be deficient in radon. A better solution is a combination of both methods, in that a pump may be used to flush water into the bottle while it is submerged, thus replacing the deficient water with fresh water sampled from the same depth. A cap may be placed on the bottle while it is being retrieved, but for a 2.5L glass jug, this should immediately be removed and a few mL of water decanted off to create the expansion buffer.

Dry the bottle and apply a label stating the date, time and source of the water.

#### 1.4.3 Setting up the equipment

The method described in this section uses the large laboratory drying unit. Other choices are discussed in Section 3.

The following should be attached to the aerator:

- Teflon Collar
- Bottle cap
- Tubing with Y-Connector
- Aerator Stones



Fig. 4 Aerator and Bottle assembly with Temperature Logger

The temperature logger, with the waterproof sensor, may be started logging the temperature any time prior to the water measurement. See the temperature logger manual. Choose a 2-minute time interval between temperature readings.

Assemble with 2" (5cm) of large, 5/16" ID vinyl tubing connecting the output of the aerator barrel to the bubble trap. Connect the other end of the bubble trap to a T-connector and thence to the screw-cap end of the large drying unit, also with 5/16" ID tubing. The other end of the large drying unit is connected, with 1/8" ID tubing, to an inlet filter mounted on the RAD7 inlet. Adaptors from 5/16" to 1/8" makes this connection easy and secure. Connect the RAD7 outlet (also with 1/8" ID tubing, adapted up to 3/16") to a T-connector and thence to the check valve on the aerator. Connect a bypass from one T-

connector to the other, with a shut-off clip on the tubing. See Fig. 3.

With the system as connected so far, set the RAD7 to purge for another few minutes. While it is purging, clamp the drying unit to the retort stand, thus supporting it about 70cm above the base, such that the cap attached to the aerator is at the right height to fit on the Big Bottle.

Press [Enter] on the RAD7 to stop purging. Then select [Setup]→[Protocol] →[Sniff] and push [Enter]. Next go to [Setup]→[Cycle] and set the cycle time to 15 minutes. Set the Format to short by choosing [Setup]→[Format]→[Short] and pushing [Enter]. Once these settings have been configured, it may be desirable to save the configuration under the User Protocol. This will make it easier to restore the necessary settings in the future. (This is described in more detail in the RAD7 manual.) Make sure the infrared printer has paper, and switch it on. Next switch off the RAD7, then switch it on again. It will print its identity and a review of the setup.

While the RAD7 is printing, remove the cap from the Big Bottle and lower the aerator stones into the water. Some water may spill during this procedure. Screw the cap attached to the aerator onto the Big Bottle. The bottle should stand comfortably on the base of the retort stand with the bubble trap and desiccant supported vertically above it. Attach the temperature sensor in direct contact with the glass of the bottle and held in place with the foam under the elastic clinching strap. Close the clip on the bypass.

#### 1.4.4 Running the test with desiccant

Once everything is set up, make sure the temperature logger is running. Once the RAD7 has finished printing out the header, go to [Test]→[Start] and push [ENTER]. The pump will start running, aerating the sample and delivering radon to the RAD7. Set the pump to run continuously by selecting [Setup]→[Pump]→[0n] and pushing [Enter]. After 15 minutes, the RAD7 will print out a short-form report. The same thing will happen again 15 minutes later, and every 15 minutes after that.

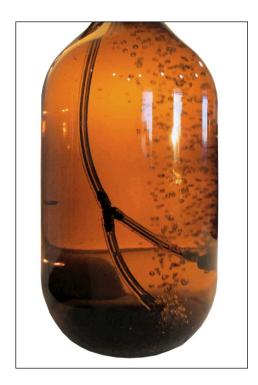


Fig. 5 Aeration in progress

Aerating the water in the Big Bottle System moves the bulk of the radon from the water into the air loop. This process may take around 45 minutes, depending on several factors.

After about 45 minutes from the start, the aeration process will be complete and the radon in the air loop will be close to equilibrium with the radon remaining in the water. The clip between the T-connectors may now be opened to allow the air flow to bypass the bottle.

At this point, it is no longer necessary to keep the pump running continuously. The pump may be set to Auto by selecting [Setup]→[Pump] →[Auto] and pushing [Enter]. To read the relative humidity go to the third status window, see section 1.4.1.

### 1.4.5 Running the test with the DRYSTIK

The 48-inch and 144-inch DRYSTIKs are both capable of bringing the RH in the RAD7 down to below 10% without the use of desiccant. The 12-inch DRYSTIK is not quite able to do that, but it brings the RH down below 20%, which gives good results if CAPTURE is used to correct for humidity. Alternatively, the 12-inch DRYSTIK may be used with a small drying tube placed between it and the RAD7 inlet. This will extend the life of the desiccant.

Before starting the test, turn the RAD7 pump off ([Setup]  $\rightarrow$  [Pump]  $\rightarrow$  [Off]  $\rightarrow$  [ENTER]). Connect the low-flow outlet of the DRYSTIK to the inlet of the RAD7 and the outlet of the RAD7 to the purge input on the DRYSTIK. Run the DRYSTIK at 100% duty cycle for an hour or more, to bring down the RH in the RAD7 and to purge it with fresh air.

Connect the system as shown in Fig. 6. What was previously (with desiccant) connected as a bypass to the aerator is now used to make the high flow output from the DRYSTIK available for rapid aeration of the sample. Once everything is set up, the DRYSTIK may be started with 100% duty cycle. This starts the aeration of the sample and the distribution of radon around the air loop. The RAD7 (with the RAD7 pump still off) should also be started. Every 15 minutes it will complete a cycle and print out the data if a printer is in position and switched on.

After 45 minutes or so (with the higher flow rate) the aeration will be complete. At this point, the clip on the bypass may be closed, thus shutting off the flow from the high-flow output (it may be necessary to put a tie-wrap around the upstream hose connections to prevent them blowing off if they are not tight). Also the duty cycle can be reduced to, say, 20% thus reducing the air flow round the loop to a sedate 0.03 L/min, which is sufficient to keep the RAD7 dry. Keep the measurement going until sufficient data points have been accumulated to reach the precision required.

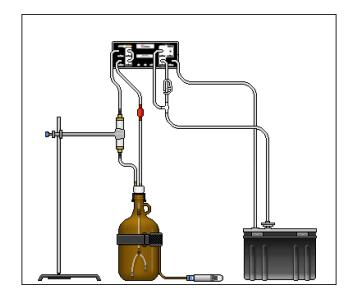


Fig. 6 Big Bottle System with DRYSTIK

## 1.4.6 Finishing the Test

If left alone, the measurement will continue indefinitely. At the end of each 15-minute cycle the RAD7 will print out the measurement of radon in air and its uncertainty. During the first 45 minutes the RAD7 will aerate the sample and respond to the growth of radon in the measurement chamber, so the cycles that are completed during this time should be ignored.

The best time to stop the measurement will depend on the level of certainty required, but it will always occur after the initial 45 minute aeration has been completed.

Radon concentrations and uncertainties are shown in the top line of the printout for each cycle. You can also look at the last reading in the second status window, by pressing [Menu] then [Enter], pressing [Enter] again and the right arrow once. Observe the uncertainty and express it as a percentage of the base value. If that percentage uncertainty is too high, calculate the ratio of this observed uncertainty to an acceptable uncertainty. The square of that ratio is the number of cycles, N, after equilibrium, that are required to reach the desired precision of measurement.

You can, if you wish, set the RAD7 to terminate the measurement at the required point automatically by setting the "Recycle" number to 'N + 2':

[Setup] → [Recycle] → ['N+2'], [Enter].

At the end of the (N+2)th cycle the RAD7 will finish the measurement and print out a run summary. Alternatively, once the (N+2)th cycle has been completed and the data printout finished, you can simply turn off the RAD7. This will terminate the measurement. The data will be stored in the RAD7 and on the printer output, but there will be no run summary printed out.

Once the measurement is terminated, unscrew the bottle cap, raise the aerator stones out of the water, close the bypass clip and set the RAD7 to purge for 8 minutes or more. This will blow water out of the stones, and also introduce fresh air into the return tubing and the RAD7.

If no more tests are to be analyzed, the equipment may now be dismantled. If there is another sample for analysis, keep the RAD7 connected as above (with the water jug removed) and purge for ten minutes. Check the relative humidity, as above, and continue the purge until the relative humidity indication in the instrument drops to 10% or below. After ten minutes, the RAD7 air outlet may be connected to the input of the drying unit by opening the bypass clip, to form a closed loop, to conserve desiccant. When the relative humidity is down to 10% or less, another test may be conducted. Repeat from 1.4.1 above.

#### 1.4.7 Interpreting the results

The RAD7 printout may appear something like Fig. 7, if the recycle number was set to N+2. Otherwise, if the measurement was terminated by switching off the RAD7, the printout will consist only of the cycles data. The run summary will be missing.

Download the RAD7 data to your computer with CAPTURE. Set the Radon Measurement Method to Big Bottle System for the automatic calculation of radon in water concentrations based on the RAD7's reported radon in air values, or set the Radon Measurement Method to Radon in Air if these values are to be calculated manually. See Section 3 for details.

In addition, there will be a temperature log stored in the Lascar temperature logger, that may be downloaded and printed with a Windows PC. Use the PC software to create a comma-delimited ASCII text file of the temperature data. For the automatic conversion of the RAD7 radon-in-air readings to radon concentration in the water sample, CAPTURE will need that temperature data.

The final step is to correct the measured value of radon in the water for decay of the radon during the time between taking the sample and analyzing it. See Section 3 for details.

DURRIDGE RAD7 Vers 2.5f 991128 Model 711 Serial 02375 Calib 29-APR-11 Last used THU 15-DEC-11 16:38 Current settings THU 15-DEC-11 16:40 Protocol: (None) Cycle: 00:10 Recycle: 07 Mode: Sniff Thoron: Off Pump: Auto Tone: Off Format: Short Units: pCi/l 1901 14.0±4.56 p Sniff THU 15-DEC-11 16:51 23.4°C RH: 7% B:6.94V 1902 92.5±10.8 p Sniff THU 15-DEC-11 17:01 23.7°C RH: 7% B:6.94V 1903 94.8±11.0 p Sniff THU 15-DEC-11 17:11 24.0°C RH: 9% B:7.03V 1904 112±11.9 p Sniff THU 15-DEC-11 17:21 24.3°C RH: 8% B:7.03V

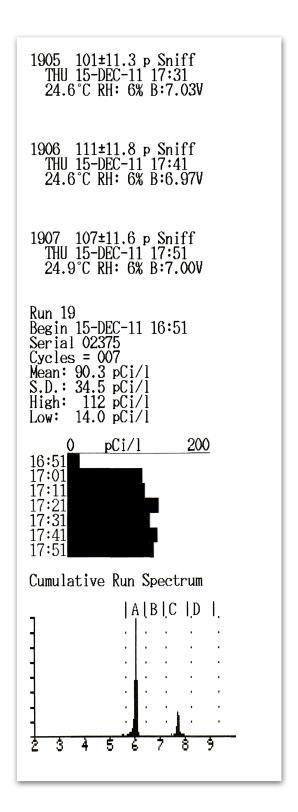


Fig. 7 Big Bottle System printout

## 2 BIG BOTTLE SYSTEM MEASUREMENT TECHNIQUE

## 2.1 The Closed Loop Concept

The RAD  $\rm H_2O$  and Big Bottle System each employ a closed loop aeration scheme whereby the air volume and water volume are constant and independent of the flow rate. The air recirculates through the water and extracts the radon until a state of equilibrium develops. The standard RAD  $\rm H_2O$  system reaches this state of equilibrium within 5 minutes, after which no more radon can be extracted from the water. The Big Bottle System needs aeration for more than 10 minutes to reach equilibrium.

With a standard RAD  $H_2O$ , the extraction efficiency of radon from the water samples is very high, and temperature independent, typically 99% for a 40 mL sample and 94% for a 250 mL sample. For the Big Bottle System, however, the extraction efficiency is temperature dependent and the temperature has to be monitored.

#### 2.2 Desiccant

The Big Bottle System is more efficient if desiccant is used at all times to dry the air stream before it enters the RAD7. A DRYSTIK model 48-ADS or 144-ADS will keep the RAD7 reasonably dry, even without any desiccant. If neither desiccant nor a DRYSTIK is used, the RAD7 will be filled with humid air and the readings will need to be corrected for humidity to obtain the correct radon concentrations. This can be done automatically using CAPTURE, which may be downloaded from the DURRIDGE web site, (http://durridge.com).

The Big Bottle System may be used with either the large drying unit, the small drying tube, a DRYSTIK, or none of these. The best solution is the DRYSTIK 144-ADS, because this keeps the RAD7 dry while minimizing the volume of the air loop.

## 2.3 Purging the System

After performing a water or air measurement, the RAD7's internal measurement chamber will continue to contain the radon that was measured. If this radon is still present in a closed air loop method, such as the Big Bottle System, when you start a new measurement the retained radon in the chamber will be added to the radon stripped from the water. To

prepare for the next water measurement, therefore, you must remove the old radon from the RAD7 and its peripherals, including the aerator heads, tubes, and desiccant. This procedure is known as "purging the system."

To purge the system, you must have a source of radon-free (or relatively radon-free) air or inert gas. For most occasions ambient air is good enough, but see below for a discussion on background. Put the RAD7 into a purge cycle with the [Test]→[Purge] command, and allow the RAD7 pump to flush clean air through the entire system for 10 minutes. Do not recycle the purge air back through the desiccant to the RAD7 until after at least 10 minutes of open-loop purging to clear out the radon.

Be sure to allow all the hoses and fittings to flush thoroughly by keeping them attached during the purge cycle for at least the first five minutes. Also be sure that the drying tube does not deplete its desiccant during the purge cycle. If the depleted (pink) desiccant gets to within 1 inch of the drying tube outlet, replace the tube with a fresh (blue) drying tube. After the first two or three minutes of purging, you may replace a small drying tube with the large laboratory drying unit, to conserve the small drying tube desiccant, and continue purging the system.

Be careful about the air you use to purge! Ambient air may not be adequately free of radon to properly prepare the system for a low level sample. The radon present in the purge air will add unwanted "background" to the next measurement. For example, a purge air radon concentration of 4 pCi/L will add about 5 pCi/L (depending on the Big Bottle System configuration) additional radon concentration to the next water result. This would be too much background to neglect when measuring samples below 20 pCi/L, but if you are measuring only water samples above 20 pCi/L, you may consider this amount of offset to be acceptable. To reduce the error due to purge air radon you may either subtract off the background from every measurement, or adopt strategies, such as use of activated charcoal during the purge, or purging with nitrogen gas or outside air, to reduce the background to acceptable levels.

One way to determine the background is to measure a "blank", a water sample containing no radon. To get radon-free water, purchase distilled water from your local pharmacy, or fill a container with tap water, and allow the container to stand closed and undisturbed for 4 weeks or more. The 4 week period allows any radon present in the water to decay away. Store your radon free water in a closed air-tight container. Remember that the background due to purge air radon will change when the air radon concentration changes, so if you intend to subtract background you should measure a blank sample at every measurement session.

An alternative method to determine background is to make a measurement of the air in SNIFF mode and note the equilibrium count rate in window A.

After purging the RAD7 with fresh air, start a measurement. After 15 minutes start a new cycle. After another ten minutes look at the count rate in Window A, in the fifth status window. From a previous printout of a water measurement, with the format set to medium or long, you can see the count rate in window A corresponding to the water radon concentration measured. Typically, for a 2.5L Big Bottle, 100 pCi/L in the water will generate about 20 cpm in window A. A background count rate of 0.5 cpm in window A (equivalent to about 2 pCi/L in air) will then produce an offset of 2.5% in the final reading.

The obvious way to reduce background is to purge with very low radon air. Outdoor air rarely exceeds 0.5 pCi/L at several feet above the ground, so you can probably get the water background to below 0.7 pCi/L by simply using outdoor air to purge. To get even lower radon air, fill a tank or balloon with outdoor air and let it age for several weeks. If you are using compressed air or inert gas, be very careful not to allow the RAD7 to pressurize, as this may cause internal damage to the pump or seals.

Another method to reduce background is to use charcoal adsorption to clean the remaining radon from the system following the purge. A small tube containing 15 grams of activated charcoal, as supplied with the system, can remove up to 98% of the remaining radon from the RAD7 when connected in a closed loop. This will reduce the system's radon to truly negligible levels for the most accurate low level radon-in-water measurement. The charcoal filter works best if you use it only after a complete purge with low radon air, which avoids overloading the

filter with radon. If the charcoal filter becomes badly contaminated it can give off some radon and actually increase the background after a purge. Store the charcoal filter with the yellow end caps installed to allow the filter to "self-clean" by waiting for adsorbed radon to decay over several weeks time. Always keep the charcoal dry by using it in conjunction with a drying tube, since water vapor can adversely affect charcoal's capacity to adsorb radon.

Even if you choose not to use elaborate methods to reduce the background, you should always purge the system between samples. It is much better to purge with ordinary room air than not to purge at all.

## 2.4 Background and Residuals

Purge air is one among several causes for background counts in the Big Bottle System. The most significant other causes are radon daughters and traces of radon left from previous measurements. The RAD7 has the unusual ability to tell the difference between the "new" radon daughters and the "old" radon daughters left from previous tests. Even so, a very high radon sample can cause daughter activity that can affect the next measurement.

After a high radon sample has been purged from the system, its decay products stay behind until they decay away. The polonium-218 isotope decays with a 3 minute half-life. In the 30 minutes following the purge, the polonium-218 decays to about a thousandth of its original activity. That still leaves a background of 100 pCi/L after a 100,000 pCi/L sample.

In addition to polonium-218, the RAD7 is sensitive to polonium-214, which can give counts for several hours after the radon has been removed. The RAD7 uses alpha energy discrimination to reject polonium-214 counts from a measurement, but a very small percentage of the polonium-214 decays slip past the discriminator. This can add background to a measurement that follows a high radon sample. The solution to the problem of daughter activity is time. Simply wait for the activity to decay away. Check the background with a blank sample. If it is still too high, keep waiting, and keep checking. The length of time you will wait depends on just how much radon your high radon sample had, and just how much background you are willing to tolerate before the next measurement. If you expect the next sample to be high also, you may want to go ahead

with the next measurement right away, considering a small amount of background acceptable.

In the case of extremely high radon samples, you may develop a background that is more persistent than daughter activity. That is possibly due to off-gassing of residual radon that has absorbed into internal surfaces. In particular, rubber and plastic parts can absorb a small fraction of the radon that passes through the system. A small fraction of a very large amount can still be significant. The radon may desorb from these materials over many hours. In the worst case you may have to allow the system to sit idle for a day or more for the absorbed radon to finish leaking out of these materials, then purge the

system again to remove the radon. A radon concentration high enough to cause a concern of this kind is very rare in natural ground water, but is possible in artificial radon sources such as radium crocks or "Revigators".

Sustained counting of very high radon concentrations can lead to the buildup of long lived lead-210 contamination of the RAD7's alpha detector. This possibility is described in the RAD7 Operator's Manual. It suffices to say that the RAD7's ability to distinguish alpha particles by energy makes it far less susceptible to the build up of lead-210 related background than other radon monitors.

## **3 CALCULATING RESULTS**

#### 3.1 How Calculation Is Made

For the standard RAD  $H_2O$ , the RAD7 uses the WAT 40 and WAT250 protocols to automate the aeration and counting process and to calculate the radon concentration in the water directly, so that the RAD7 reading is the actual radon concentration in the water.

With the Big Bottle system, however, the WAT protocols should generally be avoided. Instead, the RAD7 is used in Sniff mode to measure the radon concentration in the air loop and a conversion factor to radon in the water must be applied. The conversion factor used depends on the configuration of the Big Bottle System and on the temperature of the water at the time of the aeration. Automatic conversion is not performed by the RAD7 as in RAD H<sub>2</sub>O experiments, but is instead handled using CAPTURE. The physical configuration of the experiment needs to be specified, specifically the size of the bottle and tubing apparatus, the type of drying unit used, and the temperature of the water at the time of aeration. Without CAPTURE, this calculation may be performed manually as shown below.

### 3.2 Detailed Calculation

The basic principle is that the radon, originally in the water, is distributed around the air loop. The concentration in the air loop is measured and from this the original radon concentration in the water is calculated.

#### 3.2.1 Definitions

To begin, we need to identify all the variables involved. There are several volumes, designated with a V and a lower case suffix, total radon, Rn, radon concentrations, C, and temperature, T.

Let:

V total equivalent air volume of system

 $V_{R7}$  internal volume of the RAD7

 $V_d$  air space & pore volume of desiccant

 $V_t$  volume of tubing & aerator

V<sub>b</sub> volume of bubble trap

V<sub>h</sub> volume of head space in bottle

V<sub>a</sub> total volume of air in the system

V<sub>w</sub> volume of water in bottle

V<sub>eq</sub> equivalent air volume of water in bottle

Rn total radon in system

 $\Delta$  Rn radon lost from system

C<sub>w</sub> original radon concentration in the water

C<sub>b</sub> background radon concentration in air

C<sub>a</sub> equilibrium radon concentration in air

T temperature of water in bottle

∝ equilibrium coef. from Fritz Weigel equation

#### 3.2.2 Known Values

V<sub>R7</sub> 800mL, standard RAD7

954mL, RAD7 with oversized dome

V<sub>d</sub> 400mL, laboratory drying unit

23.7mL, small drying tube

133 mL DRYSTIK 144-ADS-3(R),

88mL, DRYSTIK 44-ADS-3(R), or

70mL, DRYSTIK 12-ADS-3(R)

V<sub>t</sub> 54mL

V<sub>b</sub> 51mL

V<sub>h</sub> 15mL

 $V_w$  2.5L

It may be noted that

- 5/16" ID tubing has a volume of 49mL/m.
- 3/16" ID is 18mL/m.
- 1/4" ID is 8mL/m.

The standard tubing set results in a total tubing volume, Vt, of 54mL.

$$V_a = V_{R7} + V_d + V_t + V_b + V_h$$

Assuming that the setup is standard,

$$V_t + V_b + V_h = 120$$
ml. Giving:

$$V_a = 120 + V_{R7} + V_d$$

With a standard RAD7 and a laboratory drying unit:

$$V_a = 120 + 800 + 400 ml = 1.32 L$$

#### 3.2.3 Head Space Loss

The head space of air above the water in the Big Bottle is there to prevent expansion of the water from breaking the bottle. However it causes a loss of radon from the sample in that the radon concentration in the head-space air will reach equilibrium with the water and, when the cap is opened, that air will escape.

If we assume a typical value for  $\propto$ , the distribution coefficient (or partition coefficient), at typical room temperature, to be 0.25, then the radon concentration in the head space will be four times the radon concentration in the water. Radon lost from the system will be:

$$\Delta Rn = (C_w \cdot V_h) / \propto$$

Care needs to be taken to ensure that the units for concentration and volume are consistent, e.g. if  $C_w$  is in Bq/m<sup>3</sup> then  $V_h$  must be expressed in m<sup>3</sup>.

### 3.2.4 Air-equivalent of water

When the radon is distributed uniformly around the air loop, some will remain in the water. The equilibrium ratio of the concentrations is  $\propto$ , as determined by the von Weigel equation. A volume  $V_w$  of water will therefore contain as much radon as a volume  $\propto V_w$  of air. This quantity,  $\propto V_w$ , may therefore be considered the air-equivalent volume of the water.

#### 3.2.5 Calculation

$$Rn = C_w \cdot V_w - \Delta Rn$$

$$Rn = C_w \cdot (V_w - V_h / \infty)$$

The total equivalent air volume of the system will be  $Va + \propto Vw$ . The total radon in the system, Rn, will be distributed around this volume, so that the concentration in the air loop at equilibrium will be:

$$C_a = Rn / (V_a + \propto V_w)$$
 or

$$Rn = C_a \cdot (V_a + \propto V_w)$$

substituting for Rn:

$$C_w \cdot (V_w - V_h/\alpha) = C_a \cdot (V_a + \alpha V_w)$$

O

$$C_w = C_a \cdot (V_a + \propto V_w) / (V_w - V_h / \propto)$$

With a standard RAD7, a laboratory drying unit and typical room temperature, such that  $\alpha = 0.25$ , this would reduce to:

$$C_w = C_a \cdot (1.29 + 0.625) / (2.5 - 0.06)$$

or

$$C_w = C_a \cdot 0.785$$

### 3.2.6 Correction for ambient/purge radon

Any radon in the purging air will simply be added to the radon in the system. Therefore:

$$Rn = C_w \cdot (V_w - V_h/\alpha) + C_h \cdot V_a$$

This modifies the equation, above, to:

$$C_w = \left[ C_a \cdot (V_a + \propto V_w) - C_b \cdot V_a \right] / (V_w - V_h / \propto)$$
 which is nearly the same as:

$$C_w = (C_a - C_b) \cdot (V_a + \propto V_w) / (V_w - V_h / \propto)$$

#### 3.2.7 Radon Graph Interpretation

The graph of radon against time generated by CAPTURE from the RAD7 data is not a horizontal straight line. There is, first, a rising slope due both to the response time of the RAD7 in Sniff mode and to the time taken to transfer radon from the water to the air. The curve goes through a small peak and then settles on a slowly descending straight line.

The intercept of this line on the Y axis, projected back to the start of the measurement, would be a good reading. However, it is not necessary to keep the measurement going long enough to determine the position and slope of that line with good accuracy. It is sufficient to average the readings from 45 minutes to between 60 and 90 minutes from the start of the measurement, depending on the radon level and the precision required.

#### 3.2.8 Decay Correction

If you collect a sample and analyze it at a later time (rather than immediately), the radon concentration will decline due to the radioactive decay. You must correct the result for the sample's decay from the time the sample was drawn to the time it was counted. If the sample is properly sealed and stored, and counted within 24 hours, then the decay corrected result should be almost as accurate as that of a sample counted immediately. Decay correction can be used for samples counted up to 10 days after sampling, though analytical precision will decline as the sample gets weaker and weaker.

The decay correction is a simple exponential function with a time constant of 132.4 hours. (The mean life of a radon-222 atom is 132.4 hours, which is the half-life of 3.825 days multiplied by 24 hours per day divided by the natural logarithm of 2.) The decay correction factor ( $D_{CF}$ ) is given by the formula

$$D_{CF} = e^{(T/132.4)}$$

in which T is the decay time in hours.

You will notice that decay times of under 3 hours require very small corrections, and you can ordinarily neglect the decay correction for samples that are collected and then quickly brought back to the lab for analysis.

To correct your result back to the sampling time, multiply it by the decay correction factor ( $D_{CF}$ ) from the chart, Fig. 9 on page 20.

## 3.3 Automatic Calculation with CAPTURE Software

Durridge's CAPTURE software for Windows and macOS makes it easy to obtain Big Bottle System radon data and automatically calculate radon in water concentrations. CAPTURE's Run Parameters window is used to specify details such as the size of the water sample and whether a DRYSTIK accessory was present during the aeration process, as shown in Fig. 8, below. The program will then automatically convert each downloaded RAD7 radon reading into a radon in water data point, using the procedure described in Section 3.2, above.

This functionality is described in depth in the Opening Data From Disk section of the CAPTURE user's manual. The CAPTURE manual is available at the Durridge website, https://durridge.com/documentation/capture\_help/.

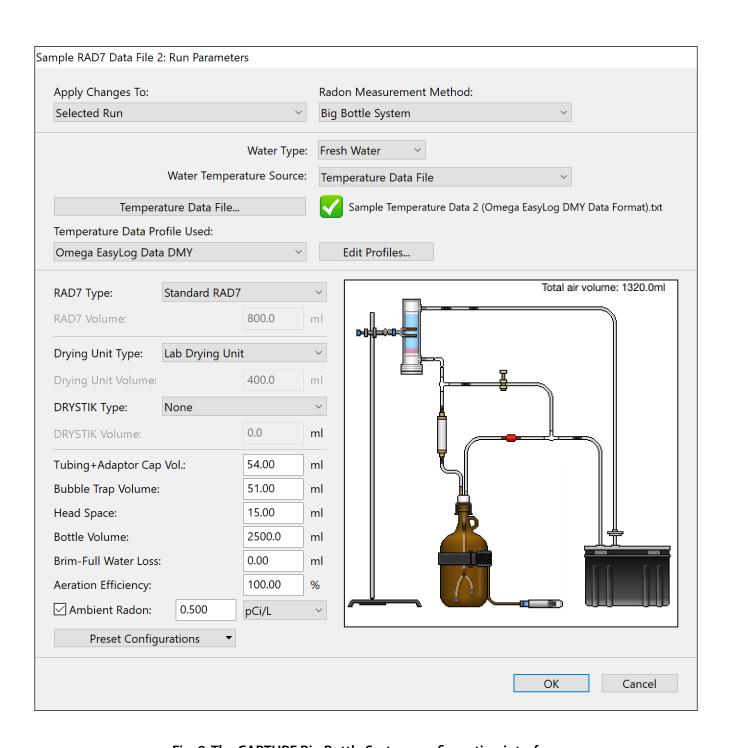


Fig. 8 The CAPTURE Big Bottle System configuration interface

Hours	DCF								
0	1.000	1	1.008	2	1.015	3	1.023	4	1.031
5	1.038	6	1.046	7	1.054	8	1.062	9	1.070
10	1.078	11	1.087	12	1.095	13	1.103	14	1.112
15	1.120	16	1.128	17	1.137	18	1.146	19	1.154
20	1.163	21	1.172	22	1.181	23	1.190	24	1.199
25	1.208	26	1.217	27	1.226	28	1.236	29	1.245
30	1.254	31	1.264	32	1.273	33	1.283	34	1.293
35	1.303	36	1.312	37	1.322	38	1.332	39	1.343
40	1.353	41	1.363	42	1.373	43	1.384	44	1.394
45	1.405	46	1.415	47	1.426	48	1.437	49	1.448
50	1.459	51	1.470	52	1.481	53	1.492	54	1.504
55	1.515	56	1.526	57	1.538	58	1.550	59	1.561
60	1.573	61	1.585	62	1.597	63	1.609	64	1.622
65	1.634	66	1.646	67	1.659	68	1.671	69	1.684
70	1.697	71	1.710	72	1.723	73	1.736	74	1.749
75	1.762	76	1.775	77	1.789	78	1.802	79	1.816
80	1.830	81	1.844	82	1.858	83	1.872	84	1.886
85	1.900	86	1.915	87	1.929	88	1.944	89	1.959
90	1.973	91	1.988	92	2.003	93	2.019	94	2.034
95	2.049	96	2.065	97	2.081	98	2.096	99	2.112
100	2.128	101	2.144	102	2.161	103	2.177	104	2.194
105	2.210	106	2.227	107	2.244	108	2.261	109	2.278
110	2.295	111	2.313	112	2.330	113	2.348	114	2.366
115	2.384	116	2.402	117	2.420	118	2.438	119	2.457
120	2.475	121	2.494	122	2.513	123	2.532	124	2.551
125	2.571	126	2.590	127	2.610	128	2.629	129	2.649
130	2.669	131	2.690	132	2.710	133	2.731	134	2.751
135	2.772	136	2.793	137	2.814	138	2.836	139	2.857
140	2.879	141	2.901	142	2.923	143	2.945	144	2.967
145	2.990	146	3.012	147	3.035	148	3.058	149	3.081
150	3.105	151	3.128	152	3.152	153	3.176	154	3.200
155	3.224	156	3.249	157	3.273	158	3.298	159	3.323
160	3.348	161	3.374	162	3.399	163	3.425	164	3.451
165	3.477	166	3.504	167	3.530	168	3.557	169	3.584
170	3.611	171	3.638	172	3.666	173	3.694	174	3.722
175	3.750	176	3.778	177	3.807	178	3.836	179	3.865
180	3.894	181	3.924	182	3.954	183	3.984	184	4.014
185	4.044	186	4.075	187	4.106	188	4.137	189	4.168
190	4.200	191	4.232	192	4.264	193	4.296	194	4.329
195	4.361	196	4.395	197	4.428	198	4.461	199	4.495
200	4.529	201	4.564	202	4.598	203	4.633	204	4.668
205	4.704	206	4.739	207	4.775	208	4.811	209	4.848
210	4.885	211	4.922	212	4.959	213	4.997	214	5.035
215	5.073	216	5.111	217	5.150	218	5.189	219	5.228
220	5.268	221	5.308	222	5.348	223	5.389	224	5.429
225	5.471	226	5.512	227	5.554	228	5.596	229	5.638
230	5.681	231	5.724	232	5.768	233	5.811	234	5.855
235	5.900	236	5.945	237	5.990	238	6.035	239	6.081

Fig. 9 Decay Correction Factors Table

## 4 ACCURACY AND QUALITY CONTROL

## 4.1 Calibration of System

The RAD H<sub>2</sub>O method relies on a fixed-volume closed-loop extraction of radon from water to air. Since the volumes are constant and the physical properties of radon are constant, we do not anticipate a need to routinely adjust the distribution (partition) coefficient. The only factors we anticipate will require "calibration checks" are sampling and laboratory technique, and the RAD7.

In sample handling you can lose a significant fraction of the radon if you do not follow consistent procedures. For this reason we recommend that you regularly review your method, and compare your results to those of other methods in side-by-side comparisons.

One way to check the accuracy of your analysis technique is to take side-by-side samples of water known to be between 100 pCi/L and 10,000 pCi/L (4,000 Bq/m³ and 400,000 Bq/m³), one sample in the Big Bottle and the other in a standard 250mL RAD  $\rm H_2O$  vial. Analyze the samples with the Big Bottle System and the standard RAD  $\rm H_2O$  system, respectively, and check to see whether the results match.

Another approach is to monitor a water supply continuously with a RAD AQUA and take a sample of that water in a Big Bottle for analysis. Please note that the RAD AQUA reading, because of its delayed response, gives the radon concentration in the water 30 minutes or more before the reading is given. Thus when a sample for the Big Bottle is taken, the user should wait at least 30 minutes and perhaps an hour before taking the latest RAD AQUA reading. This precaution is only necessary if the radon concentration in the water is changing.

As part of your quality assurance plan, you should regularly check the RAD7 for its ability to determine radon in air, and periodically send the RAD7 in for a check-up and recalibration. Government agencies usually recommend or require annual or bi-annual recalibration of radiation measurement instruments. You can find more information about calibration in the RAD7 Operation Manual.

DURRIDGE recommends against the use of radium-226 solutions in any RAD H<sub>2</sub>O or Big Bottle System due to the risk of permanent contamination.

## 4.2 Accuracy and Precision

A number of factors affect the accuracy and precision of a radon in water measurement. Most critical among these factors is the sampling technique, which was discussed in greater detail in a previous section. Other factors include the sample concentration, sample size, counting time, temperature, and background effects.

### 4.2.1 Sampling Technique

Similar samples may vary by as much as 20% due to uneven aeration, losses during sample acquisition and transfer and/or variation of the water being sampled, between earlier and later samples. By paying very careful attention to detail it may be possible to get the variation down to under 5%.

### 4.2.2 Sample Concentration

You can usually determine high concentrations with a better precision than low concentrations (when the precision is expressed in terms of percent uncertainty). This is because a higher concentration gives a greater number of counts per minute, yielding more favorable counting statistics. If the concentration is too high, however, you may exceed the upper limit of the RAD7's linear range.

#### 4.2.3 Sample Size

A larger sample size gives a greater number of counts per minute, and so improves sensitivity and precision at low radon concentrations. But the larger sample size reduces the method's range, and increases temperature effects. Thus the Big Bottle System is intended primarily for measuring low radon concentrations, such as may be encountered in open water or some domestic water supplies.

#### 4.2.4 Counting Time

Longer counting times improve sensitivity and precision by accumulating a greater total number of counts, which gives more favorable counting statistics. With the Big Bottle System, the counting

time is virtually unlimited. However, factors such as the ambient air radon concentration and other uncertainties limit the value of counting times beyond about two hours.

#### 4.2.5 Temperature

The effect of temperature on the calculation of radon concentrations in 40mL vials is negligible, and for 250mL vials it only becomes significant at temperatures outside 15 - 25 deg C (60 - 77 deg F). For the Big Bottle System however it is necessary to know the temperature of the water in the bottle at the time of aeration to within +/- 1 deg C (2 deg F). The temperature logger supplied with the equipment is designed to provide this information by measuring the temperature of the glass about halfway up the bottle.

The equilibrium ratio,  $\propto$  of radon concentration in water to air is given by:

$$\alpha = 0.105 + 0.405 \, \cdot \, e^{\left( \text{-}0.0502 \, \bullet \, \text{T} \right)}$$

Where T °C is the temperature of the air-water interface (Weigel, 1978).

At typical room temperature  $\propto$  is about 0.25, making the air-equivalent volume of the water about 0.6L.

#### 4.2.6 Relative Humidity

Provided the desiccant does not get hydrated during the measurement, or if an Active DRYSTIK is used, the relative humidity in the RAD7 may be kept low during the entire one or more hours of the measurement. In any case, CAPTURE may always be used to correct the readings, automatically, for high humidity.

### 4.2.7 Background Effects

By careful attention to details, one can reduce the background in any RAD  $\rm H_2O$  system to insignificant levels. We previously discussed how to control the background due to purge air radon content and residual radon and its progeny. The uncontrollable, or "intrinsic", background of the RAD7 is low enough to ignore in all but the most demanding cases. The intrinsic background of the RAD7 is less than 1 count per hour, corresponding to a 40 mL water sample concentration of less than 2 pCi/L (0.3 pCi/L for the 250 mL sample and eight times lower still in the Big Bottle System). In principle, you can achieve a

background this low if you completely eliminate all radon and progeny from the system before aerating the water, but that will require a fair amount of effort and patience. A more realistic background to shoot for, using the Big Bottle System, would be between 0.2 and 0.5 pCi/L (between 8 and 20 Bq/m³) in the water. If you know the background well enough, you can subtract it from the reading and have reasonable confidence in the result.

# 4.3 Comparison of RAD H₂O and Big Bottle System with Other Methods

Fig. 10 is a table of typical numbers to give a basis for comparison. Some laboratories may be able to get better results with the RAD  $H_2O$  than this table indicates, while others may not. The precision figures include counting statistics only, with no adjustment for sampling variation or decay of the sample.

Note that standard laboratory analysis often entails a long delay between sampling and analysis, which can significantly increase the error and raise the detection limit (DL) and the lower limit of detection (LLD). Also note that the background figure used to calculate the RAD H<sub>2</sub>O and Big Bottle System precision, DL, and LLD is conservatively estimated to reflect typical field usage. The most demanding and patient operator should be able to reduce background to less than 0.02 cpm (rather than the 0.10 cpm given in the table), which will allow for DL's and LLD's lower than those listed.

## 4.4 Quality Assurance

A proper quality assurance plan should follow the guidelines set by the USEPA as described in [Goldin]. Compliance with certification programs certainly require an approved quality assurance plan.

The elements of a quality assurance plan include blank samples, duplicate samples, and spiked samples. Often, the plan provides for blind samples to be measured in an inter-comparison program. If a quality control measurement deviates beyond the acceptable range, the operator must cease to make measurements until the cause of the deviation has been discovered and corrected. Therefore, the quality assurance plan should specify the range of acceptable measurement deviations, often in the form of a "control chart". The operator should maintain complete records of the quality control measurements and their deviations.

Method	RAD H <sub>2</sub> O 40	RAD H <sub>2</sub> O 250	Big Bottle System	Liquid Scintillation	Lucas Cell			
Sample Size (mL)	40	250	2500	10	10			
Sensitivity (cpm/pCi/L)	0.008	0.05 0.3		0.09	0.05			
Background (cpm) 0.1*		0.1*	0.1*	15	0.25			
2-sigma uncertainty at 300 pCi/L (in pCi/L)								
20-minute count	88	35		32	35			
60-minute count 51		20	2.5	19	20			
120-minute count	36	14	1.8	14	14			
2-sigma uncertainty at 100 pCi/L (in pCi/L)								
20-minute count	53	20		24	20			
60-minute count 31		12	1.5	14	12			
120-minute count 22		8.5	1.1	10	8.5			
DL (C=2*(1+sqr(1+2*B)) in pCi/L) (NPDWR 40-CFR-41.25)								
20-minute count	40*	6*		28	9			
60-minute count 19*		3*	0.4*	16	4			
300-minute count	7*	1*	0.18*	7	2			
LLD (C=4*(1+sqrB)) in pCi/L (Altshuler)								
20-minute count	60*	10*		41	13			
60-minute count	29*	5*	0.6*	23	6			
300-minute count	11*	2*	0.25*	10	3			

Fig. 10 Method Comparison

<sup>\*</sup> Note that the RAD7 background is conservatively estimated to be 0.1 cpm to arrive at these figures. With care an operator should be able to reduce the background to less than 0.02 cpm, which would significantly lower the RAD7 DL and LLD figures from those displayed.

## 5 CARE, MAINTENANCE and TROUBLESHOOTING

## 5.1 Warning on Pump Direction

The Big Bottle system cannot tolerate the reversal of the air connections at the aerator head or the RAD7. A check valve should be used at all times to prevent the possibility of sucking water into the RAD7, should a connector be accidentally reversed. If a reversed connection occurs, the check valve prevents the water from rising past the aerator head by blocking its path. Do not allow the RAD7 to continue pumping against a blocked check valve, as this may cause damage to the pump or to the RAD7's internal seals.

## 5.2 Warning on Tipping the Aeration Unit

Use the large retort stand to support the bubble trap and desiccant in a vertical position, with the bottle and aerator directly beneath it.

Never operate the Big Bottle System in any position other than upright! If it tips to any direction it may allow water to pass through the outlet tube toward the RAD7. If liquid water reaches the RAD7, it could permanently damage critical internal parts, resulting in an expensive repair bill.

If water ever enters the RAD7, or if the RAD7 ever goes swimming in the water, it will probably cease to operate and immediate steps should be taken to minimize the impact on the instrument.

Keep the RAD7 upright. This will prevent water from touching the detector, which is close to the face plate at the top of the dome. Put a piece of tubing on the RAD7 outlet with the other end in a sink. Use the RAD7 pump if it still works or, otherwise, an external pump into the inlet, to blow air through the instrument. When water ceases to be blown out of the outlet, put desiccant upstream of the RAD7 to dry out the air path. When the air path is fully dry (after dry air has been blown through it for approximately one hour), remove the face plate from the case, empty the water out of the case and blow dry the case and the RAD7 electronics.

Once there is no visible water in or on the instrument, it can be put in an oven at 50°C for a few hours to dry out completely. Additionally, desiccated air can be passed through the air path until the air leaving the RAD7 drops below 10% RH. After this

treatment further corrosion will be prevented, and the RAD7 will boot once more and you can use the internal RH sensor to measure how dry the air path is. At this point the instrument should be returned to DURRIDGE for service.

### **5.3 Stones Maintenance**

After performing many radon in water measurements, the stones may begin to show stains or even begin clogging due to the buildup of mineral deposits. If the mineral buildup is light and low in radium content, we see no reason for concern. Heavy deposits may be removed from the stones by soaking it in an acid solution, followed by a thorough rinse with clean water. Replacement stones are readily available at any aquarium shop.

## 5.4 High Humidity

With cycles longer than 5 minutes, the pump normally stops after the first five minutes. This is insufficient to aerate the sample thoroughly. The solution, after the measurement has started, is to set the pump to ON [Setup]→[Pump]→[0n] [Enter] and leave it running for the entire first cycle. After that, the bypass clip can be opened, allowing the air flow to bypass the water, and the pump set to AUTO, which would pump for five minutes at the start of every cycle. This will keep the humidity low throughout the measurement.

Without desiccant and without an Active DRYSTIK, the pump will run continuously on AUTO and can be turned off after 15 minutes of aerating the sample by setting the pump to OFF. The data should be downloaded with CAPTURE and displayed with the "show corrected radon" button checked.

With an Active DRYSTIK, the RAD7 pump should be set to OFF and the DRYSTIK pump used to circulate air round the loop. After 45 minutes of pumping and aerating the sample, the bypass clip can be opened and the duty cycle of the DRYSTIK changed to, say, 10%. The relative humidity in the RAD7 may not be as low as with desiccant and it may still be appropriate to click the "show corrected radon" in CAPTURE.

## 5.5 Foaming

While clean water causes no problem, some natural waters contain foaming agents that will cause bubbles to rise up out of the aerator. The bubble trap supplied with the Big Bottle System is designed to limit the upward migration of the foam. This arrangement makes it difficult for bubbles to rise up as far as the desiccant and reduces the concern about foaming.

## 5.6 Plastic Bottle Denting

Large soda bottles with insufficient wall strength may become dented inward during aeration. Any indentation will reduce the volume of the bottle and displace water upward towards the desiccant. For this reason bottles with strong walls that can resist denting are recommended. This problem is less likely to affect soda bottles with volumes of less than 2L. Glass bottles are not affected. The risk of this problem occurring may be affected by the strength of the RAD7's internal pump. To decrease the effective pump strength, add one or more additional inlet filters to the RAD7.

## 5.7 Glass Jug Care

As mentioned above, it is essential to leave a small air pocket in the jug when it is sealed to prevent smashing the glass when the water inside heats up. CAPTURE provides automatic compensation for this head space in the calculation of radon in the water.

To protect the jug from accidental damage when carrying it around, protective shoulder bags are available. Please see the Durridge web site (www.durridge.com) for details.

## 5.8 Technical Support

It is not expected that the Big Bottle System will need maintenance or service in normal use. For technical support please contact service@durridge.com, or call Durridge Company at (978) 667-9556.

## **6 BACKGROUND TO RADON-IN-WATER MEASUREMENT**

#### 6.1 About Radon-in-Water

Radon originates from the radioactive decay of naturally occurring uranium and radium deposits. These elements can be found, in trace amounts, in almost all soils and rocks. Being a gas, radon can escape from mineral surfaces and dissolve in ground water, which can carry it away from its point of origin. We rarely find radon in high concentrations in surface waters, due to its rapid dispersal into the atmosphere.

High concentrations of groundwater radon prevail in parts of New England, New Jersey, Maryland, Virginia, and the mountainous western states of the U.S. Typical groundwater sources average between 200 and 600 pCi/L of radon. Roughly 10 percent of public drinking water supplies have concentrations of over 1,000 pCi/L, and around 1 percent exceed 10,000 pCi/L. Smaller water systems appear to be disproportionally affected by high radon. [Milvy, EPA]

Radon was first noticed in water supplies by J.J. Thomson, a pioneer in the science of radioactivity, in the first decade of the 1900s. [Hess, Frame] At first, scientists and doctors believed radioactivity to have benign, even curative, effects on the human body. Early research linked high radon concentrations to natural hot springs long thought to have miraculous powers. But eventually, science came to understand the dangers of radiation exposure, after a number of serious accidents and fatalities. [Caulfield]

In the 1950s airborne radon decay products emerged as the probable cause of high incidences of lung cancer among underground mine workers. Study of environmental radioactivity revealed unusually high groundwater radon concentrations in the vicinity of Raymond, Maine. [Bell] In the 1960s, scientists began to investigate the effect of ingested and inhaled radon gas, observing the uptake of radon by digestive organs and its dispersal through the bloodstream. [Crawford-Brown] By the 1970s, radon was widely recognized as a major component of our natural radiation exposure. By the late 1970s, Maine had initiated a program to attempt to reduce public exposure to waterborne radon, having discovered cases in which groundwater concentration exceeded 1 million pCi/L. [Hess]

Federal action on the problem of radon in drinking water picked up in the 1980s with a nationwide program to survey drinking water supplies for radioactivity and to assess the risk to public health. Congress directed the Environmental Protection Agency (EPA) to take action on radioactivity in drinking water, and in 1991 the EPA officially proposed a Maximum Contaminant Level (MCL) for radon in public drinking water of 300 pCi/L. This MCL may one day become binding on public water supplies. [Federal Register, EPA]

## 6.2 Health Risks Due to Waterborne Radon

Waterborne radon leads to health risk by two pathways: inhalation of radon and its decay products following the release of radon gas from water into household air, and the direct ingestion of radon in drinking water.

The risk of lung cancer due to inhaled radon decay products has been well documented through the study of underground mine workers. The cancer risk due to ingestion, primarily cancer of the stomach and digestive organs, has been estimated from studies of the movement of radon through the gastrointestinal tract and bloodstream. Radon has not been linked to any disease other than cancer. The cancer risk from the inhalation pathway probably far exceeds that from the ingestion pathway. [Crawford-Brown, Federal Register]

In a typical house, with typical water usage patterns, a waterborne radon concentration of 10,000 pCi/L will yield an average increase to indoor air concentrations of about 1 pCi/L. The 10,000:1 ratio, while not to be considered a hard rule, has been verified through theoretical models and empirical evidence. [Hess] In a house with a high radon in water content, air radon concentrations tend to rise dramatically with water usage, especially in the vicinity of the water-using appliance, but decline steadily after the water usage tails off. [Henschel]

In most houses, waterborne radon is a secondary source of indoor radon, far exceeded by soil gas infiltration. It is an exception, though not a rare one, that waterborne radon is the major contributor to elevated radon in air. A homeowner who has

discovered elevated air concentrations, and whose house uses private well water, should test the water for radon content to assess the water's contribution to the airborne radon. This test ought to be done before any attempt to mitigate soil gas infiltration, particularly if other wells in the area have been found to have radon. [Henschel]

## 6.3 Physical Properties of Waterborne Radon

Radon gas is mildly soluble in water. But, being a gas, it is volatile. It tends to leave the water upon contact with air. This is known as aeration.

The rate of radon transfer from water to air increases with temperature, agitation, mixing, and surface area. In household water usage, showers, baths, dishwashers, laundries, and toilets all provide adequate aeration to release a high percentage of the water's radon content into household air. [Prichard]

In principle, the radon will continue to be released from water as the aeration process continues, until a state of equilibrium develops. According to Henry's Law of dilute solutions, equilibrium will occur when the water concentration and air concentration reach a fixed ratio for a certain temperature. This ratio, derivable from the Henry's Law constant for radon dissolved in water, is known as the distribution coefficient or partition coefficient.

For radon in water at 20 degrees C (68 F) the distribution coefficient is about 0.25, so radon will continue to release from the water until the water concentration drops to about 25 percent of the air concentration. Remember that as the radon leaves the water into the air it raises the air concentration and lowers the water concentration. At lower temperatures the distribution coefficient increases, rising to 0.51 at 0 degrees C (32 F). At higher temperatures the distribution coefficient decreases, dropping to about 0.11 at 100 degrees C (212 F). An empirical expression for the distribution coefficient of radon in water as a function of temperature can be found in [Weigel].

# 6.4 Radon as a Tracer for Groundwater movement

Soil and rock typically contain significant concentrations of uranium and radium. Radon is continually being created in the ground so that

groundwater often has high radon content. By contrast, open water contains very little dissolved radium. That, together with the proximity of the water surface, means that the background concentration of radon in sea and lake water far from land is very low.

Radon, then, with its 4-day half life, is an almost perfect tracer for measuring and monitoring the movement of ground water into lake and sea water along the shore [Lane-Smith, Burnett].

In open water, fast-response continuous radon measurement at high sensitivity is provided by the RAD AQUA [www.durridge.com]). For ground water it is usually more convenient to use the RAD H<sub>2</sub>O, but this is not sensitive enough for open water. The gap between the RAD AQUA and the standard RAD H<sub>2</sub>O is filled by the Big Bottle System. This has one tenth the sensitivity of the RAD AQUA and is eight times more sensitive than a standard RAD H<sub>2</sub>O.

# 6.5 Standard Methods for Radon in Water Analysis

Several methods have been developed to measure radon in water. Three of these are Gamma Spectroscopy (GS), Lucas Cell (LC) and Liquid Scintillation (LS).

Gamma spectroscopy seeks to detect the gamma rays given off by radon's decay products from a closed container of radon bearing water. While simple in concept, this method lacks the sensitivity to detect radon at the lower levels now considered important.

The Lucas Cell method has been in use for decades for laboratory analysis of radon-222 and radium-226 (via radon emanation). The LC method tends to be somewhat labor intensive, using a complicated system of glassware and a vacuum pump to evacuate a Lucas (scintillation) cell, then bubble gas through the water sample until the cell fills. The cell is then counted by the usual technique. In the hands of a skilled technician this method can produce accurate, repeatable measurements at fairly low concentrations. [Whittaker, Krieger (Method 903.1)]

The Liquid Scintillation method dates to the 1970s. A liquid scintillation cocktail is added to the sample in a 25 mL glass LS vial. The cocktail draws the radon out of the water, so that when it decays the alpha particles scintillate the cocktail. The method uses standard LS counters, which are highly

automated and can count several hundred samples in sequence without intervention. The EPA has determined that the LS method is as accurate and sensitive as the LC method, but less labor intensive, and less expensive. [Prichard, Whittaker, Hahn (Method 913.0), Lowry, Vitz, Kinner, Hess]

In comparison with the above, the RAD  $\rm H_2O$  offers a method as accurate as LS but faster to the first reading, portable, even less labour intensive and less expensive. It also eliminates the need for noxious chemicals. The Big Bottle variant of the RAD  $\rm H_2O$  has the same advantages as the standard RAD  $\rm H_2O$  together with an 8-fold increase in sensitivity, giving it a lower limit of detection that is an order of magnitude better than liquid scintillation.

## 6.6 Mitigation Strategies

Two main strategies have emerged for the removal of radon from water. Both of these are applicable to point-of-entry (POE) water treatment in residences and small public water supplies.

Granular Activated Carbon (GAC) attempts to filter the water by adsorbing radon on a charcoal bed that holds onto the radon until the radon decays. GAC systems can be effective and relatively inexpensive for residential use, but can create new problems. As the radon and its progeny decay in the GAC column, they give off gamma radiation. The gamma radiation may be a health concern to residents when the influent radon concentration is high, the GAC column is poorly shielded for high energy radiation, and the residents are likely to spend significant periods of time in the radiation field. Over time, a long lived decay product, lead-210, builds up in the column, which may pose disposal problems in systems with moderate to high radon concentrations in the influent. For that reason GAC is most often recommended for influent concentrations of up to around 5,000 pCi/L. GAC maintenance is simple and inexpensive, and the GAC bed has an expected service life of 5 to 10 years. [Henschel, Lowry, Rydell]

Aeration brings water into contact with a stream of low radon air, which strips the radon from the water, then exhausts the radon bearing air to the atmosphere. Aeration systems offer effective removal of radon without the buildup of gamma radiation or waste material, but tend to be substantially more expensive than GAC to install and maintain in a residential setting. Aeration can be used over the entire range of influent concentrations, though very high influent concentration may require a multiple stage system to reduce the effluent concentration to acceptable levels. [Henschel, Lowry, NEEP]

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## DURRIDGE Company Inc. 900 Technology Park Drive Billerica, MA 01821

Telephone: (+1) 978-667-9556
Fax: (+1) 978-667-9557
Web: www.durridge.com
Email: service@durridge.com

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