THESIS

TRITIUM UNCERTAINTY ANALYSIS FOR SURFACE WATER SAMPLES AT THE SAVANNAH RIVER SITE

Submitted by

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ABSTRACT

TRITIUM UNCERTAINTY ANALYSIS FOR SURFACE WATER SAMPLES AT THE SAVANNAH RIVER SITE

Radiochemical analyses of surface water samples, in the framework of Environmental Monitoring, have associated uncertainties for the radioisotopic results reported. These uncertainty analyses pertain to the tritium results from surface water samples collected at five locations on the Savannah River near the U.S. Department of Energy's Savannah River Site (SRS). Uncertainties can result from the field-sampling routine, can be incurred during transport due to the physical properties of the sample, from equipment limitations, and from the measurement instrumentation used. The uncertainty reported by the SRS in their Annual Site Environmental Report currently considers only the counting uncertainty in the measurements, which is the standard reporting protocol for radioanalytical chemistry results. The focus of this work is to provide an overview of all uncertainty components associated with SRS tritium measurements, estimate the total uncertainty according to ISO 17025, and to propose additional experiments to verify some of the estimated uncertainties. The main uncertainty components discovered and investigated in this paper are tritium absorption or desorption in the sample container, HTO/H_2O isotopic effect during distillation, pipette volume, and tritium standard uncertainty. The goal is to quantify these uncertainties and to establish a combined uncertainty in order to increase the scientific depth of the SRS Annual Site Environmental Report.

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To my wife and son, I appreciate your sacrifices to allow me to complete this endeavor. You mean everything to me.

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Chapter 1 Introduction

The Savannah River Site (SRS) is a mostly restricted area of approximately 300 square miles that is located in Aiken, Allendale and Barnwell Counties of South Carolina. The southern boundary boarders the Savannah River for approximately 40 miles. The site is owned by the US Department of Energy (DOE) and is managed and operated by the Savannah River Nuclear Solutions. It has been used for nuclear weapons material processing since the 1950's. The Savannah River Site stopped producing tritium on site in 1988. Today the site is the only site with the capability of extracting tritium from current nuclear weapons and recycling it to maintain the nation's stock pile. Tritium is made at commercial nuclear power stations and then sent to the Savannah River Site for extraction, purification and then to be put back into the nuclear the weapons. The site is managed by Savannah River Nuclear Solutions, LLC. The majority of the radionuclide releases into the Savannah River are in the form of very mobile tritiated water (SRS Environmental Report 2009). Tritium is the ^{3}H isotope of hydrogen with one proton and 2 neutrons. It readily combines with oxygen to form "tritiated water." Most tritium in water samples consists of one ^{3}H (T), one ^{1}H, and one oxygen atom, often abbreviated HTO (MARLAP 2004). The HTO form was assumed for this study. The current environmental monitoring program at SRS involves continuous monitoring of surface water for quantification of many radionuclides including tritium. Tritium accounts for most of the liquid discharges from the SRS liquid effluents. This paper will focus on the tritium measurement uncertainty from the time a river water sample is taken from the river until it is counted in the Savannah River National Laboratory (SNRL). An aerial map of SRS together with the dedicated sampling sites for tritiated water is shown in Figure 1.

SRS has surface water sampling locations at five points in the Savannah River and additional sampling locations on each of the major streams that run through the site and empty into the Savannah River. SRS performs a tritium balance comparison at various site streams and Savannah River monitoring locations annually. The tritium balance is compared either through 1) direct releases plus measured shallow groundwater migration of tritium, 2) tritium transport measured in SRS streams, or 3) river transport measured downriver of SRS after subtraction of any contribution above SRS. The river transport method was the primary focus of this study. SRS has a sampling point upstream of the site for the collection of background samples. There are sample locations before and after the Vogtle Electric Generating Plant (VEGP) to allow for subtraction of the nuclear plant's contribution to the HTO in the river. Finally, the last sampling location is a few miles past the site. SRS can subtract the VEGP contribution and the background as obtained from the upstream sampling location from the last sampling location down river from the site to obtain the SRS tritium releases.

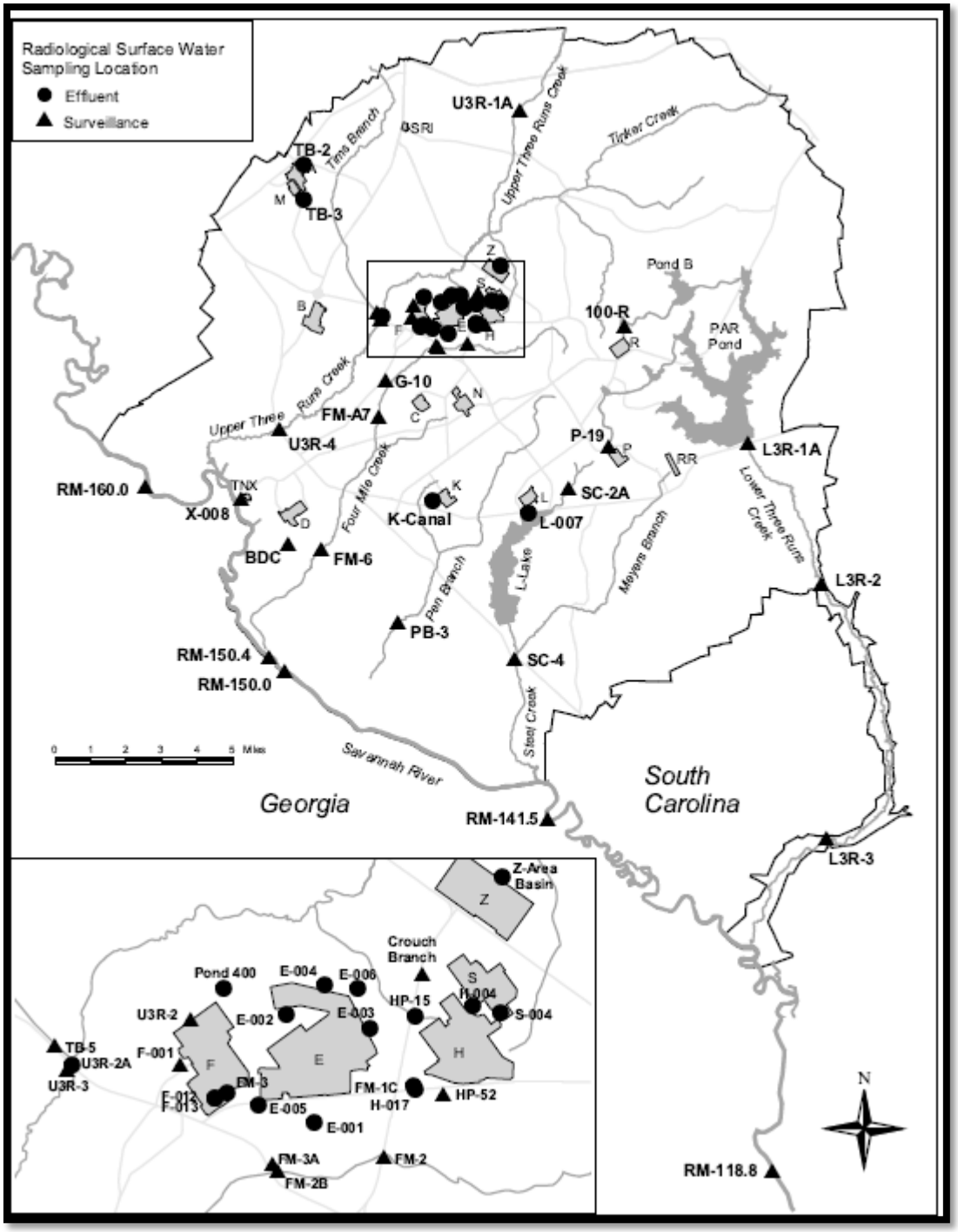

Figure 1. Radiological Surface Water Sampling Locations.

All measurements have associated uncertainties. Uncertainties can result from the sampling routine, the samples' physical properties during transport, equipment limitations, and the statistics of the sample measurements. The goal is to estimate the total tritium activity

concentration uncertainty for each of the sampling sites reported in the SRS Environmental Report. The motivation behind this uncertainty calculation is compliance with ISO/IEC 17025 (ISO 2005). It requires that calibration and testing laboratories have and apply procedures for estimating measurement uncertainties. This International Standard by the International Organization for Standardization (ISO) references the Guide to the Expression of Uncertainty in Measurement (GUM 1995) for details.

SRS reports the measurement results from the samples collected at different points in units of curies per liter, maximum contaminant levels, minimum detection limit, and a dose to the maximum exposed individual. This thesis focuses on the activity concentration measured. As international standardization requires the use of SI units for physical measurements, and as many US professional bodies endorse this standardization, the activity concentrations measured in the course of this study are expressed in units of becquerel (Bq) per gram which translates directly into units of Bq per mL. Traditional units may be presented in parenthesis for some results.

Figure 2. The weekly 10 L composite sample collection and ISCO sampler calibration at River Mile 160 (RM-160)

Every week, a composite sample of approximately 10 L is collected in a 15 L plastic container from each station by sampling 30 mL every 30 minutes (Figure 2). In the laboratory, a 300 mL aliquot is extracted from each of the five stations' 15 L composite sample containers, and the tritium concentration is determined by Liquid Scintillation Counting (LSC). Before counting, the 300 mL sample is distilled to remove any contaminants from the raw sample that could degrade the measurement quality. The first 2-5 mL of the distilled sample are discarded. The next 25 mL are collected and a 10 mL aliquot is pipetted into a plastic liquid scintillation vial together with 12 mL of PerkinElmer Liquid Gold™ LLT scintillation cocktail. After dark adaptation, this vial is counted for 8 hours in a LSC. The overall process is shown in the diagram in Figure 3. The LSC software segregates the counts into bins according to energy. Only the counts below the maximum beta energy of Tritium (18 keV) are counted.

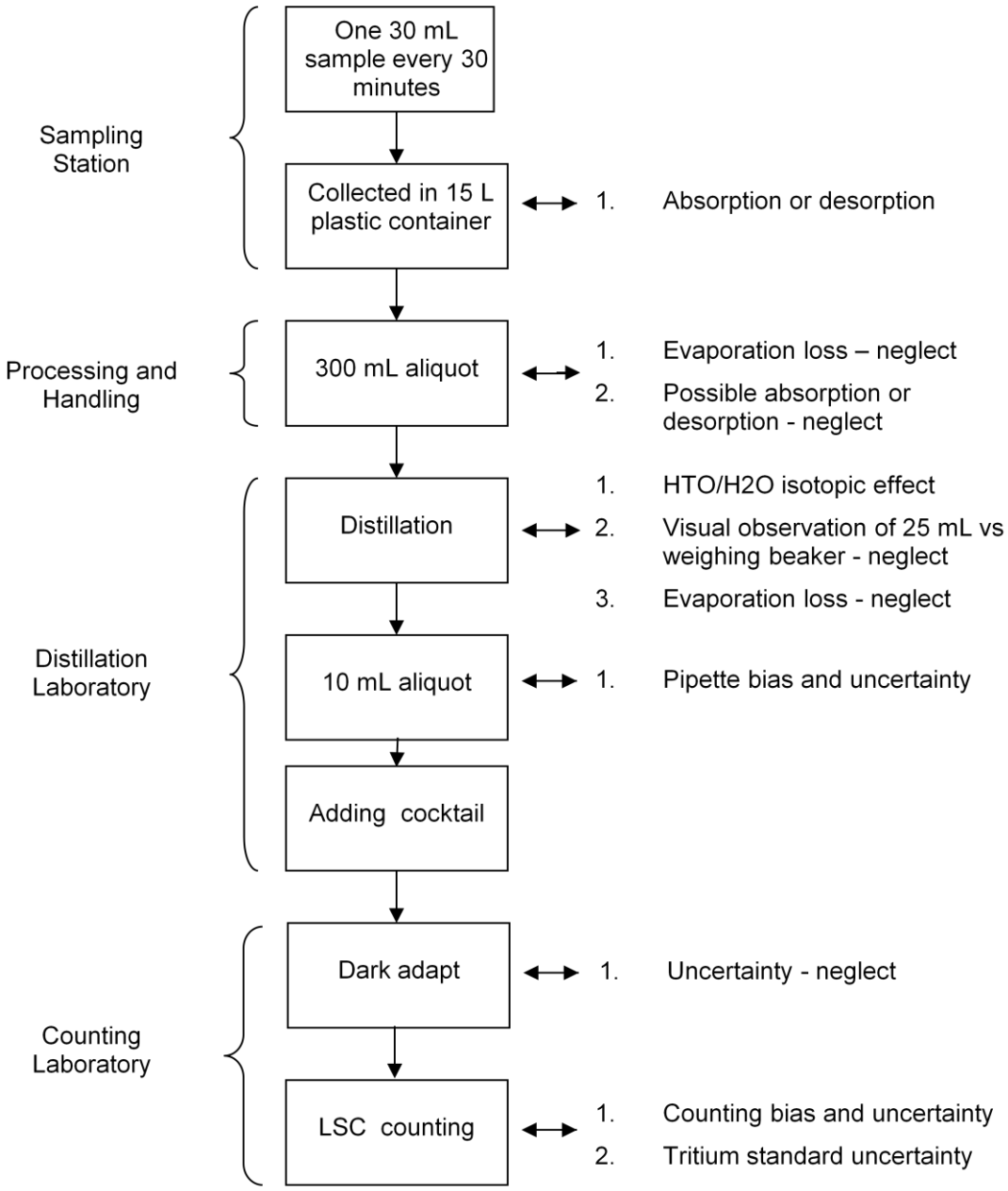

Figure 3. SRS tritium sampling and analysis process for the river monitoring program.

The reason for this specific sampling system is that each sampler takes 336 samples per week. This provides a composite average of the tritium concentration in the river with 52 evenly spaced samples per year. However, the sampling system does not allow for determination of the

absolute maximum, minimum, and variances of individual data. An analysis of the statistics of the composite sampling method is beyond the scope of this thesis.

The uncertainty contributions studied in the course of this project include only contributions after the sample water enters the 15 L container. All uncertainty estimates were obtained by using either available SRS data or results from measurements performed at the Colorado State University (CSU) laboratory. The available data included uncertainty contributions due to the pipetting and the weekly SRS tritium measurements. Other uncertainties had to be tested, such as the uncertainties associated with the distillation process and those due to absorption of tritium by the sample containers. Equations for the total uncertainty were derived by combining quantified uncertainties according to the error propagation formula (Knoll 2010, Tsoulfanidis 2011).

The design of the experiments made use of a few basic assumptions. Individual uncertainty contributions are deemed uncorrelated. Tritium is always assumed to be evenly distributed in the sample (Imboden 1977). Thus, when an aliquot is taken, the activity concentration of the aliquot is the same as the activity concentration in the original standard solution. Uncertainty contributions due to the delay before counting, age of the cocktail, and storage temperatures of the sample vials with cocktail are deemed negligible. This appears reasonable because SRS always counts a known standard which is prepared together with the samples. SRS and CSU use different LSC's, which could be a source of additional variation between the measurements performed at CSU and SRS. The differences between the two machines performing LSC analyses will be discussed later. The uncertainties associated with the CSU experiments were derived separately.

Uncertainty contributions due to the calibration standard, counting statistics, and pipetting are estimated from the manufacturer, the 2009 SRS weekly environmental data, and the available pipette calibration data, respectively. The effects of tritium absorption into the sample container and the HTO/H_2O isotopic effect during distillation were investigated by conducting experiments at CSU.

Chapter 2 Theory

a) Types of Uncertainties

The term uncertainty depends on its adjectives (GUM 1995). The term alone means doubt. Without its adjectives, the term could mean both qualitative and quantitative uncertainties. Uncertainty evaluations can be grouped into two types, Type A and B uncertainty evaluations. A Type A evaluation is based on the statistical analysis of the data by determining the standard deviation of the mean of the measured quantity, by least squares fit to the data, or by using analysis of variance (ANOVA). A Type B evaluation is based on scientific judgment supported by previous data, experience and knowledge, manufacturer's specifications, calibration data, etc. Type B evaluation was only used qualitatively in this work to reject some uncertainty contributions.

b) Standard Deviation

The standard deviation is the most common method for representing uncertainties. The definition of the standard deviation follows the definition provided in the literature (GUM 1995, Tsoulfanidis 2011). All data distributions in this thesis are assumed Gaussian. The standard deviation is representative of the dispersion of data about the mean. The probability of a randomly chosen data point to be within one standard deviation of the mean is 68%. The probability of a randomly chosen data point to be within two standard deviations of the mean is 96%. A standardized representation of the number of standard deviations is provided by the k-value. In this thesis, two standard deviations are used in tables and error bars on graphs to represent the expanded probabilities for individual data points. The uncertainty calculations are performed using one standard deviation.

The experimental standard deviation is sometime misrepresented or used interchangeably with the term standard error of the mean (GUM 1995). The standard deviation does not represent error, such as mistakes in the conduct of the experiment, it represents the dispersion of the measured values about a true mean.

c) Propagation of Uncertainties

The methodology to combine various uncorrelated uncertainty contributions for the measured variables is outlined is derived from equation 2.1 (GUM 1995, Tsoulfanidis 2011, Knoll 2010). Combining of uncertainties is termed the law of propagation of uncertainty (GUM 1995), also called the error propagation formula in other documentation (Knoll 2010, Tsoulfanidis 2011).

$$\sigma_{\bar{f}} = \sqrt{\sum_{i=1}^{N}\left(\frac{\partial f}{\partial \bar{x}_i}\right)^2 \sigma_i^2} \quad \text{for} \quad \bar{f} = f\left(\bar{x}_1, \bar{x}_2, \bar{x}_3, \ldots \bar{x}_N\right) \tag{2.1}$$

where σ_T is total combined uncertainty, N is the number of individual parameters, f is the function combining the input parameters, the x_i represent the individual parameters, and σ_i is estimated uncertainty on each parameter.

If the individual input parameters appear in terms of a sum or a difference, the combined uncertainty is derived according to:

$$f(x_1, x_2) = a_1 x_1 \pm a_2 x_2$$

where a_1 and a_2 are constants.

$$\sigma_T = \sqrt{a_1^2 \sigma_1^2 + a_2^2 \sigma_2^2}$$

If $a_1 = a_2 = 1$, then

$$\sigma_T = \sqrt{\sigma_1^2 + \sigma_2^2} \tag{2.2}$$

If the individual input parameters appear in terms that are multiplied, the combined uncertainty is derived according to:

$$f(x_1,x_2) = ax_1x_2$$

$$\frac{\partial f}{\partial x_1} = ax_2$$

$$\frac{\partial f}{\partial x_2} = ax_1$$

$$\sigma_T = a\sqrt{x_2^2\sigma_1^2 + x_1^2\sigma_2^2}$$

$$\frac{\sigma_T}{f} = \frac{a\sqrt{x_2^2\sigma_1^2 + x_1^2\sigma_2^2}}{ax_1x_2} = \sqrt{\left(\frac{\sigma_1}{x_1}\right)^2 + \left(\frac{\sigma_2}{x_2}\right)^2} \qquad (2.3)$$

If the individual input parameters appear in terms that are divided, the combined uncertainty is derived according to:

$$f(x_1,x_2) = a\frac{x_1}{x_2}$$

$$\frac{\partial f}{\partial x_1} = a\frac{1}{x_2}$$

$$\frac{\partial f}{\partial x_2} = -a\frac{x_1}{x_2^2}$$

$$\sigma_T = a\sqrt{\frac{\sigma_1^2}{x_2^2} + \frac{x_1^2\sigma_2^2}{x_2^4}}$$

$$\frac{\sigma_T}{f} = \frac{a\sqrt{\frac{\sigma_1^2}{x_2^2} + \frac{x_1^2\sigma_2^2}{x_2^4}}}{ax_1/x_2} = \sqrt{\left(\frac{\sigma_1}{x_1}\right)^2 + \left(\frac{\sigma_2}{x_2}\right)^2} \qquad (2.4)$$

If the individual input parameters appear in terms of multiplication and division, the combined uncertainty is derived according to:

$$f(x_1,x_2,x_3) = \frac{ax_1x_2}{x_3}$$

$$\frac{\partial f}{\partial x_1} = a\frac{x_2}{x_3}$$

$$\frac{\partial f}{\partial x_2} = a\frac{x_1}{x_3}$$

$$\frac{\partial f}{\partial x_3} = -a\frac{x_1x_2}{x_3^2}$$

$$\sigma_T = a\sqrt{\frac{x_2^2\sigma_1^2}{x_3^2} + \frac{x_1^2\sigma_2^2}{x_3^2} + \frac{x_1^2x_2^2\sigma_3^2}{x_3^4}}$$

$$\frac{\sigma_T}{f} = \frac{a\sqrt{\frac{x_2^2\sigma_1^2}{x_3^2} + \frac{x_1^2\sigma_2^2}{x_3^2} + \frac{x_1^2x_2^2\sigma_3^2}{x_3^4}}}{a\frac{x_1x_2}{x_3}} = \sqrt{\frac{x_2^2\sigma_1^2}{x_3^2}\left(\frac{x_1x_2}{x_3}\right)^{-2} + \frac{x_1^2\sigma_2^2}{x_3^2}\left(\frac{x_1x_2}{x_3}\right)^{-2} + \frac{x_1^2x_2^2\sigma_3^2}{x_3^4}\left(\frac{x_1x_2}{x_3}\right)^{-2}}$$

$$\frac{\sigma_T}{f} = \sqrt{\left(\frac{\sigma_1}{x_1}\right)^2 + \left(\frac{\sigma_2}{x_2}\right)^2 + \left(\frac{\sigma_3}{x_3}\right)^2} \tag{2.5}$$

d) SRS Uncertainty Equation

The SRS weekly reported activity concentration for each sampling site in the Savannah River is derived according to the following equations:

$$C_{sample\_reported} = \frac{C_{sample-blank}}{C_{standard-blank}}C_{standard} \tag{2.6}$$

$$C_{sample-blank} = C_{sample\_measurement} - C_{blank\_measurement} \tag{2.7}$$

$$C_{standard-blank} = C_{standard\_measurement} - C_{blank\_measurement} \tag{2.8}$$

where $C_{sample\_reported}$ is the reported activity concentration of a sample from one of the sample sites, $C_{sample-blank}$ is the measured activity concentration of the sample minus the measured activity concentration of the blank, $C_{standard-blank}$ is the measured activity concentration of the standard minus the measured activity concentration of the blank, and $C_{standard}$ is the known activity of the calibration standard.

During the data analysis and the experiments, a set of biases was identified and estimated. A correction factor (*CF*) provides the inverse to a bias and is combined according to

$$CF = CF_{pipette} \times CF_{HTO\_Isotopic\_Effect} \times CF_{Absorption} \times CF_{Desorption} \quad (2.9)$$

where $CF_{pipette}$ is the pipette correction discussed in Chapter 4, $CF_{HTO\_Isotopic\_Effect}$ is the HTO/H_2O isotopic effect correction discussed in Chapter 5, and $CF_{Absorption}$ and $CF_{Desorption}$ are the correction factors from absorption and desorption in the 15 L composite sample container discussed in Chapter 6. Considering these correction factors, the final sample results should be calculated and reported as:

$$C_{sample\_reported} = \frac{C_{sample-blank} \times CF}{C_{standard-blank}} C_{standard} \pm \frac{\sigma_{total}}{C_{sample\_measurement}} \% \quad (2.10)$$

where $\frac{\sigma_{total}}{C_{sample\_measurement}}$ is total relative uncertainty on the measured sample.

The blank and known concentration vials are prepared in the SRS laboratory according to the same protocol as the sample taken from the river. The blank activity concentration measurement ($C_{blank\_measurement}$) is subtracted from the sample and standard activity concentration

measurements to account for machine and cocktail background. The relative uncertainty $\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)$ on the activity concentration in the blank sample vial is calculated according to:

$$\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)^2 = \left(\frac{\sigma_{pipette}}{m_{pipette}}\right)^2 + \left(\frac{\sigma_{blank\_counts}}{n_{blank}}\right)^2 \tag{2.11}$$

where $\sigma_{blank\_measurement}$ is the uncertainty on the blank counting measurement , $\sigma_{pipette}$ is the standard deviation of the mass measurement during pipette calibration , $m_{pipette}$ is the average mass measurement during pipette calibration, and $\left(\frac{\sigma_{blank\_counts}}{n_{blank}}\right)$ is the relative statistical counting uncertainty.

The relative uncertainty, $\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)$, on the activity concentration in the sample is calculated according to:

$$\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2 = \left(\frac{\sigma_{pipette}}{m_{pipette}}\right)^2 + \left(\frac{\sigma_{sample\_counts}}{n_{sample}}\right)^2 \tag{2.12}$$

where $\sigma_{sample\_measurement}$ is the uncertainty on the measured sample, $C_{sample\_measurement}$ is the activity concentration of the measured sample, and $\left(\frac{\sigma_{sample\_counts}}{n_{sample}}\right)$ is the relative statistical counting uncertainty on the measured sample.

The blank is subtracted from the measured sample activity concentration. Uncertainties of measurements that are added or subtracted must be converted back to absolute values in order to combine according to:

$$\sigma^2_{sample-blank} = C^2_{sample\_measurement}\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2 + C^2_{blank\_measurement}\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)^2 \quad (2.13)$$

where $\sigma_{sample-blank}$ is the uncertainty on the net sample activity concentration.

The relative uncertainty, $\left(\frac{\sigma_{standard\_measurement}}{C_{standard\_measurement}}\right)$, on the activity concentration measurement in the calibration standard is calculated according to:

$$\left(\frac{\sigma_{standard\_measurement}}{C_{standard\_measurement}}\right)^2 = \left(\frac{\sigma_{pipette}}{m_{pipette}}\right)^2 + \left(\frac{\sigma_{standard\_counts}}{n_{standard}}\right)^2 \quad (2.14)$$

where $C_{standard\_measurement}$ is the activity concentration in the standard as counted at CSU, and $\left(\frac{\sigma_{standard\_counts}}{n_{standard}}\right)$ is the relative statistical counting uncertainty on the measured standard.

The blank is subtracted from the measured standard activity concentration. Uncertainties of measurements that are added or subtracted must be converted back to absolute values in order to combine according to:

$$\sigma^2_{standard-blank} = C^2_{standard\_measurement}\left(\frac{\sigma_{standard\_measurement}}{C_{standard\_measurement}}\right)^2 + C^2_{blank\_measurement}\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)^2 \quad (2.15)$$

where $\sigma_{standard-blank}$ is the uncertainty on the net standard activity concentration.

The total relative uncertainty, $\left(\frac{\sigma_{total}}{C_{sample\_measurement}}\right)$, on the activity concentration in the sample is calculated according to:

$$\frac{\sigma_{total}}{C_{sample\_measurement}} = \sqrt{\left(\frac{\sigma_{standard-blank}}{C_{standard-blank}}\right)^2 + \left(\frac{\sigma_{CF}}{C_{CF}}\right)^2 + \left(\frac{\sigma_{absolute\_standard}}{\mu_{absolute\_standard}}\right)^2 + \left(\frac{\sigma_{sample-blank}}{C_{sample-blank}}\right)^2} \quad (2.16)$$

where $\left(\frac{\sigma_{total}}{C_{sample\_measurement}}\right)$ is the final combined uncertainty in percent for the sample measurement. $\left(\frac{\sigma_{CF}}{C_{CF}}\right)$ is the relative uncertainty on the HTO/H_2O effect discussed in Chapter 5, and the $\left(\frac{\sigma_{absolute\_standard}}{\mu_{absolute\_standard}}\right)$ relative uncertainty is reported by the manufacturer.

d) Triple-to-Double Coincidence Ratio (TDCR)

The CSU LSC uses the TDCR method to calculate the efficiency of the LSC. This method does not require a known calibration standard to calculate the efficiency of the machine. Use of the TDCR allows for the measurement of a pure beta-emitting radionuclide to within +/- 10% uncertainty (Cassette and Bouchard 2012). For certain radionuclides, this method might provide more accurate results than the classical LSC method of comparing samples to a known standard, where standard uncertainties of greater than 10% are common. The TDCR approaches the value of the efficiency defined by the classical method as quenching agents are decreased. The TDCR method requires three photo multiplier tubes situated at 120 degree angles from each other in close proximity to the sample vial. Two UV photons are measured in double coincidence if they are detected within the set coincidence time in two separate photomultiplier tubes. Three UV photons are measured in triple coincidence if they are detected within the set coincidence time in the three separate photomultiplier tubes. The TDCR estimates the efficiency according to

$$eff \approx TDCR = \frac{\text{triple coincidence counts}}{\text{double coincidence counts}} \tag{2.17}$$

The number of counts registered in the instrument is divided by the time in minutes to yield counts per minute (cpm). The nuclear disintegrations in the sample per minute (dpm) is given by the measured counts per minute divided by the TDCR. Since the experiments were either distilled or used DI water, the TDCR uncertainty should be less than the reported maximum of +/-10%. The TDCR method was used for all experiments in this study. As all of the results in this study are compared to similar results provided by the same instrument, the bias on the TDCR cancels.

e) Optimizing Region of Interest (ROI)

The ROI was optimized by determining the maximum figure of merit (FOM) for the instrument. The FOM equation is given by

$$FOM = \frac{\text{efficiency}^2}{\text{background}} \tag{2.18}$$

where the efficiency is given in percent, and the background is given in cpm. The FOM was used to optimize the regions of interest (ROI) for three concentrations (0.16, 0.37, 0.67 Bq/g) used in the experiments. By calculating the FOM for different ROI's, the optimized ROI for 0.16 Bq/g (4.5 nCi/L, approximately 2.5 times background) is from channels 62 to 171. The optimized ROI for 0.37 Bq/g (9.9 nCi/L) is from channels 60 to 171. The optimized ROI for 0.67 Bq/g (18 nCi/L) is from channels 60 to 169. These ROI's are valid through the entire time of the study because the FOM only has to be re-determined for a certain analysis if the cocktail is changed, instrument is moved, or a PMT is replaced (EPA 2012).

f) t-test

The t-test is used to determine if two numbers are statistically different and not just different due to the random variations of the measurement. The t-test provides the number of combined standard deviations between the two central values. If two separate distributions are

assumed, the value calculated by equation 2.19 provides a measure of the probability that the two central values are statistically different. For this thesis, measurements with t-values greater than 1.5 will be considered statistically different. The equation for the t-test is

$$t\text{-}value = \frac{|\mu_1 - \mu_2|}{\sqrt{\sigma_1^2 + \sigma_2^2}} \tag{2.19}$$

where μ is the mean of the two distributions.

Chapter 3 Materials

a) Calibration Standards

The calibration standard used in all of the experiments was an Eckert & Ziegler Analytics (EZA, Atlanta, GA) SRS 86114-154 500 mL liquid in Flame Reagent Bottle, 9308 Bq (0.25 µCi) +/- 20% in 500.28 g H_2O. This standard was prepared gravimetrically from a master solution. The master solution was calibrated by Eckert & Ziegler by liquid scintillation counting. The Eckert & Ziegler calibrating sources are National Institute of Standards and Technologies (NIST) traceable through a measurements assurance program described in USNRC Regulatory Guide 4.15, Rev 1, February 1979 and compliant with ANSI N42.22-1995.

Tritium has a half life of 12.3 years. The reference date of the standard used is 12/5/2011. The activity is adjusted for decay by equation 3.1.

$$A = A_0 e^{-\lambda t} \tag{3.1}$$

where A = decay corrected activity (Bq (Becquerel))

A_0 = initial activity (Bq)

λ = decay constant (Eq 3.2)

t = time since initial activity

$$\lambda = \frac{\ln(2)}{T_{1/2}} \tag{3.2}$$

where $T_{1/2}$ is the half life.

b) Liquid Scintillation Counter

All tritium measurements were performed using a LabLogic 300SL Liquid Scintillation Counter with TDCR Technology (Hidex, Turku, Finland). The interface software is MikroWin™ (Mikrotech, Germany). Running the LSC counter creates two electronic files. The

first file contains all necessary information on the configuration of the LSC during the run. The second file contains the raw data. An Excel spreadsheet with macros provided by the vendor facilitates the import and manipulation of the raw data, including a data analysis and a graphic routine. The instrument specifications are shown below in Table 3.1.

Table 3.1: LabLogic LSC instrument specifications.

Energy range Betas	0-2,000 keV
Energy range Alphas	0-10,000 keV
Efficiency	^{3}H unquenched up to 70% in the open window
(E^2/B) = figure of merit	^{3}H (8 mL water sample, 12 mL IN-FLOW) > 80 in the optimized window
Efficiency	^{3}H (8 mL water sample, 12 mL IN-FLOW) > 26% in the optimized window
Background	^{3}H (8 mL water sample, 12 mL IN-FLOW) < 9 cpm in the optimized window

Chapter 4 Uncertainties Calculated from Available Data

The data for some uncertainty contributions were readily available from SRS. However, the distillation and the absorption and desorption uncertainties required additional experimental tests to establish their contributions to the total uncertainty.

a) SRS Pipette Uncertainty

SRS pipettes used for the volumetric measurement of sample aliquots are calibrated monthly. The calibration is performed by 10 independent mass measurements, where the average deviation from the true value is not permitted to exceed +/- 0.5% to pass. The most current standard deviation available to the author was from June 2011. The average pipette mass was 10.01789 g which is an average bias of 0.178 %. The standard deviation was 0.0146 g. The relative standard deviation was obtained by dividing the standard deviation by the average pipette mass yielding 0.145%.

b) SRS Counting Uncertainties

To obtain a value for the total estimated uncertainty, the relative statistical counting uncertainty must be calculated for the weekly sample, the blank, and the calibration standard. The relative counting statistical uncertainty is the square root of the total number of counts collected over the entire counting interval divided by the number of counts.

Chapter 5 Methods

a) CSU LSC Instrument and Background Uncertainty Experiment

After some testing, it became evident that the total uncertainty for the experiments was too small to explain the variation of the CSU LSC measurements. An attempt was made to retrospectively compensate for the instrument variation by the repeated measurement of the activity concentration in three known standard solutions (0.16, 0.67, and 18.5 Bq/g). These independently obtained experimental results are termed instrument uncertainty. The three known concentrations were prepared the same way as all the other experiments with approximately 12 mL of Ultima Gold LLT (PerkinElmer Life & Analytical Sciences B.V., Groningen, The Netherlands). They were also dark adapted for 10 hours. They were counted 30 times for 1800 seconds. The relative standard deviation of the activity concentration was used to estimate the relative instrument uncertainty.

b) Distillation Correction Factor and Uncertainty Estimation Experiment

Two scales were used for mass measurements. The FisherScientific (Denver, CO) A-160 serial # 19983 was calibrated on December 30, 2011. This scale was used for any mass measurement below 50 g which included vial measurements and small beakers used to add small quantities of the tritium standard to other larger containers. The other scale was the Fisher XL400D serial # 3866 which was calibrated on December 30, 2011. The Fisher XL400D was used for measurements between 50 and 400 g which included filling the 1 L standard dilutions and the distillation boiling flask. The FisherScientific A-160 and Fisher XL400D uncertainties and associated biases are calculated in Appendix A.

The purpose of the distillation experiments is to determine the correction factor (CF) and uncertainty for the HTO/H_2O isotopic effect. The boiling point of HTO is 100.76 °C compared to 100.00 °C for H_2O (NCRP 1979). The difference between the two boiling points is due to the differences between the HTO and H_2O masses. H_2O weighs 18 atomic mass units (AMU) while HTO weighs 20 AMU. Since HTO weighs 11% more than H_2O, HTO has a lower vapor pressure and a higher boiling point. When distilling liquids into separate aliquots, the first aliquots will show a higher concentration of the lower mass molecules than the original solution. The distillation setup is shown in Figure 5.1 below.

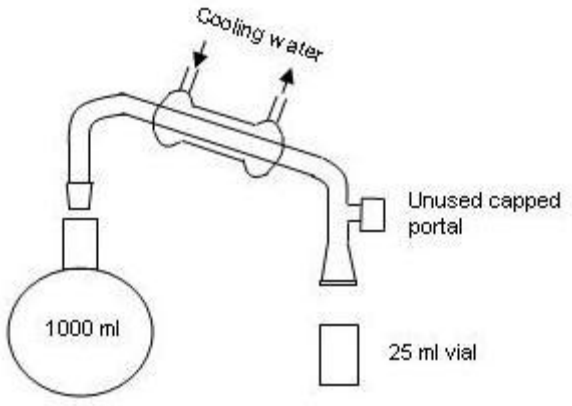

Figure 5.1: The SRS distillation setup

The distillation experiment setup at CSU was the same as the SRS distillation setup except the 1000 mL boiling flask had a larger extra port for adding the sample and cleaning. This extra port could impact the recovery by collecting distillate. All the vials were weighed before and after the distillation process to obtain a recovery percentage. The recovery percentages were recorded for each distillation experiment. The recovery percentage accounts for all distillate loss which includes the condesate on the cleaning port, condesate on any other glass surface, and evaporation loss.

The distillation method tested was the method described in the SRS technical reference manual L3.23 (SRS Determination of Tritium (HTO) in Water 2010). To summarize the major points, approximately 300 mL of the sample water is boiled (Fig 3.1). According to the SRS protocol, the first 2 - 5 mL are discarded. The next 25 mL are collected. Of that 25 mL sample, an approximately 10 mL aliquot is prepared with 12 mL Ultima Gold in a plastic LSC scintillation vial. The sample is dark adapted for greater than 10 hours and counted for 2.5 hours.

The goal of this experiment was to determine the relative activity concentration recovery of the first 25 mL and its uncertainties compared to the known original concentrations shown in Table 5.1. This experiment was performed at two distinct tritium concentrations (0.16 and 0.67 Bq/g). The initial concentration standards are approximately twice and ten times the background. The CSU trials distilled the entire 300 mL into 13 or 14 25-mL sections in 29 mL glass vials. The standard aliquots were prepared and stored in 1 L glass containers.

Table 5.1: 1 L distillation standard dilutions

Distillation Sequence Number	Activity Goal
6	0.16 Bq/g
5,7,8	0.16 Bq/g
9	0.16 Bq/g
10,11,12	0.67 Bq/g

An important difference in this experiment from the SRS process is the temperature of the boiling water. Due to the lower atmospheric pressure at CSU, the boiling water temperature at CSU is 97 °C compared to 100 °C at SRS. Effects from the differences in boiling temperature and humidity were not investigated in further detail. Another difference was the CSU boiling flask which included a cleaning port. The port collected condensate during distillations. It will be assumed that the condensate exhibits the same activity concentration as the original standard

solution thus it would have no effect on the recovery percentage of the first 25 mL vial. Since all distillation mass recoveries were greater than 97.7 % for all distillations, effects due to the condensate on the cleaning port will be assumed to be negligible.

Both, SRS and CSU used the same type of burners (Electromantle, Essex, England) with heat settings 1 to 10. Even though the boiling temperature is determined by atmospheric pressure, the heat settings could be varied from 1 (lowest heat) to 10 (highest heat). The heat settings were varied in the experiments to determine if the vigorousness of the boil had any effect on the experimental results. Any heat less than heat setting #8 was deemed ineffective due to the longer boiling time. Distilling at the heat setting #8 instead of heat setting #10 approximately doubled the distillation time in Distillations 10,11, and 12 compared to Distillations 5 and 6.

c) Absorption and Desorption Correction Factor and Uncertainty Estimation Experiments

Tritium has been found to absorb into hydrogenous materials such as rubber and plastics (MARLAP 2004, Dickson 1990). These materials can be contaminated to deep levels such that it is very difficult to completely remove tritium from their volume. The published literature does not describe appropriate correction procedures for absorption or desorption. However, the activity concentration in various vials and cocktail mixtures as a function of time described in a previous study (Barquero and Arcos 2000) closely corresponds to the results obtained in the course of this study. The previous authors attributed the observed activity concentration degradation to cocktail degradation rather than absorption. This study is aimed at providing further information by examining the activity concentration in a set of glass and polyethylene vials using a single cocktail over an extended period of time.

Absorption effects may be important for the analysis of the SRS water samples as they are transferred and stored in five separate containers during sample collection and preparation prior

to counting. Three of these containers are glass and two are plastic. Data on the SRS containers and estimated sample storage times are presented in Table 5.2. As absorption in glass is generally deemed negligible (MARLAP 2004), this study will quantify absorption or cocktail instability effects in both plastic and glass.

Table 5.2. Types of sample containers and estimated storage times.

Container	Approximate Stay Time	Material	Uncertainty Importance
15 L Composite Sample	0-2 weeks	Nalgene	Investigated
300 mL Aliquot	15 hours	Glass	Neglected
1000 mL Boiling Flask	1 hour	Glass	Neglected
25 mL Sample Vial	20 minutes	Glass	Neglected
20 mL LSC Vials	3.5 to 10 hours	Plastic	Investigated

The two most common types of polyethylene, HDPE and LDPE, have a chemical composition of $(C_2H_4)^n$ (Peacock 2000). Tritium can easily exchange with any hydrogen atom in a surrounding medium (NCRP 1979). Given the chemical composition of polyethylene, tritium exchange and absorption are expected to be measurable. The type of glass used in laboratories is usually Borosilicate glass. The majority of its chemical composition is silica (SiO_2), boron oxide (B_2O_3), sodium oxide (Na_2O), and several minor additives (Glassco Glassware 2012). The exchange and trapping of free tritium atoms in the lattice structure of the glass appears to be of minor importance. Absorption in glass should be much less than in polyethylene due to the reduced number of hydrogen atoms. Glass vials were used as controls to observe absorption in plastic vials and the establishment of an appropriate correction factor for the observed count rate reduction as a function of time.

Approximately 12 g of Ultima Gold LLT were added to Perkin Elmer part #6000477 20 mL plastic and to the glass vials and weighed. The manufacturer of the glass vials is unknown. Approximately 10 g of standard solution of three different activity concentrations (0.16 Bq/g,

0.67 Bq/g, and 18.6 Bq/g) were added to the vials. The vials were shaken and weighed. After approximately 10 hours of dark adapting, the vials were counted six consecutive times for 9000 s. This time duration was chosen to reduce the average relative counting uncertainty of the lowest activity concentration to 1.4%.

i) 20 mL Vial Absorption

The 20 mL polyethylene liquid scintillation vials and caps used by SRS are COMAR, part #12-0052-002 (Buena, NJ), and Wheaton, part #239231 (Millville, NJ). These exact parts could not be procured by CSU. Perkin Elmer 20 mL polyethylene vials, part #6000477(Waltham, MA), were used instead. The Perkin Elmer vials are coated with a micron thin PTFE®-type coating on the inside surface which is designed to reduce the diffusion of classical type solvents. This coating may also reduce the diffusion rate of tritium with respect to the diffusion rate in SRS vials.

For the test, approximately 12 mL of Ultima Gold LLT cocktail were added to the three plastic and three glass vials and weighed. Approximately 10 mL of the standard solution of three different activity concentrations (0.16 Bq/g, 0.67 Bq/g, and 18.6 Bq/g) were added to the vials and weighed. The vials were shaken approximately 10 times and dark adapted for more than 10 hours. The vials were counted periodically over a 64 day period.

ii) 1000 mL Bottle Absorption

Nine 1000 mL Nalgene Thermo Scientific wide-mouth bottles composed of high density polyethylene (HDPE), part# 2104-0032 lot 1051029 (Rochester, NY), were tested to determine if absorption in the 15 L bottles during the two week sampling period would produce an observable effect. The 15 L composite sample bottles used by SRS are Nalgene number 4 Low Density Polyethylene (LDPE). The difference between HDPE and LDPE is not expected to influence

absorption characteristics significantly. In addition, the SRS sampling container is filled by 30 mL samples every 30 minutes rather than at once at the onset of the experimental investigation. As the test sample has more contact time with the plastic for absorption to occur, experimental results are expected to be conservative with respect to actual absorption in the SRS sampling containers.

iii) 1000 mL Bottle Desorption

The same nine 1000 mL Nalgene bottles used in the two week absorption test were subsequently emptied and rinsed. The bottles were dried lightly with a paper towel. After drying, CSU DI water was added. An additional aliquot of the DI was counted to establish the sample blank for this particular test. A sample was drawn from each bottle after one week and counted. The nine bottles used in the one week desorption test were then again emptied, rinsed, and dried lightly with a paper towel. The procedure described above was repeated; however, some of the samples collected during this process had to be counted after some delay.

Chapter 6 Results and Discussion

a) LSC Instrument Uncertainty

Three activity concentration levels and two different vial types were counted six times each. The instrument variation was relatively low, ranging from 0.02 to 0.002. The data collected are displayed in Appendix B. Data from Appendix B are averaged for comparison in Tables 6.1 and 6.2. The uncertainties decreased as the activity concentration increased. This trend is expected because the higher concentrations have lower relative counting uncertainties. Since instrument uncertainties are greater than the non instrument uncertainties for low activity concentrations, a protocol for periodic checks on the instrument seems to be warranted. Non instrument uncertainty was the average relative statistical counting uncertainty of the six measurements and the scale uncertainty combined in quadrature.

Table 6.1. Instrument uncertainty data for 20 mL plastic vials.

20 mL Vial Concentration (Bq/g)	Relative Instrument Uncertainty (k=1)	Relative Non instrument Uncertainty (k=1)
0.16	0.022	0.015
0.67	0.011	0.007
18.6	0.002	0.004

Table 6.2. Instrument uncertainty data for 20 mL glass vials.

20 mL Vial Concentration (Bq/g)	Relative Instrument Uncertainty (k=1)	Relative Non instrument Uncertainty (k=1)
0.16	0.015	0.012
0.67	0.014	0.007
18.6	0.005	0.001

The instrument uncertainty does not depend on the scale or background uncertainties because the same vial and activity concentration is measured every time.

b) Distillation Correction Factor and Uncertainty Estimation

The distillation correction factor was determined by performing 12 distinct distillations. Due to standard dilutions which resulted in activity concentrations within the range of the blank samples, the first four distillations had to be discarded. The activity concentrations for Distillations 5 to 9 were approximately two times background. The activity concentrations for Distillations 10-12 were approximately ten times background. When it was noticed that the activity concentration for Distillations 5 to 9 was approximately two times background, the activity concentration was raised to approximately ten times background to follow recommendations in the standard literature (ISO 1999).

Distillations 5 and 6 were boiled on the highest heat setting (#10) during the entire distillation. Distillations 7, 8, and 9 were boiled on the highest heat setting (#10) until the first distillate was collected in the sample receptacle, then the heat was turned down to a lower setting (#8). Distillations 10-12 were heated at a lower setting (#8) for the entire distillation time. The distillation data are shown in Appendix C.

The results of the distillations are grouped into three sets according to the different heat settings and the activity concentration recovery is graphed versus the total mass recovered in Figures 6.1-6.3. The graphs generally follow a similar pattern, starting with the first activity concentration appearing to overestimate the actual activity concentration, followed by a minimum recovery value, and a subsequent slow increase. All of the distillation curves in Figures 6.1-6.3 demonstrate the isotopic effect by showing that heavier HTO is delayed till the later distillation receptacles except for the first activity concentration as discussed later in this section. The isotopic effect is most important in the first 25 mL as those are eventually counted and reported in the SRS environmental report.

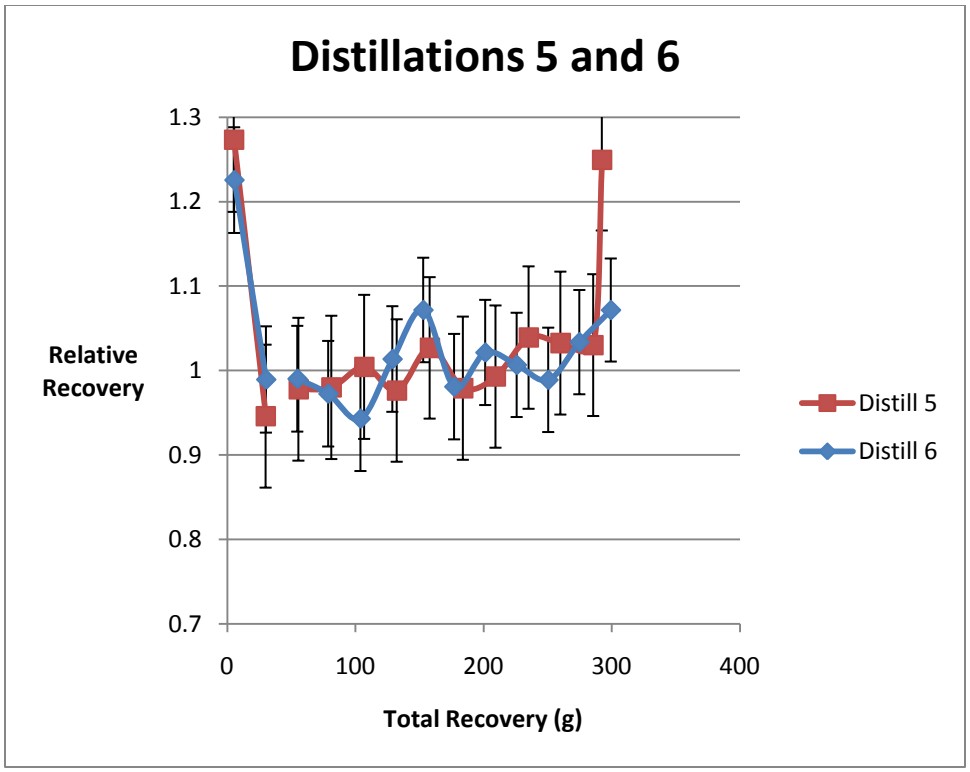

Figure 6.1: Distillations heated on the highest setting the entire time, uncertainties are reported at k=2.

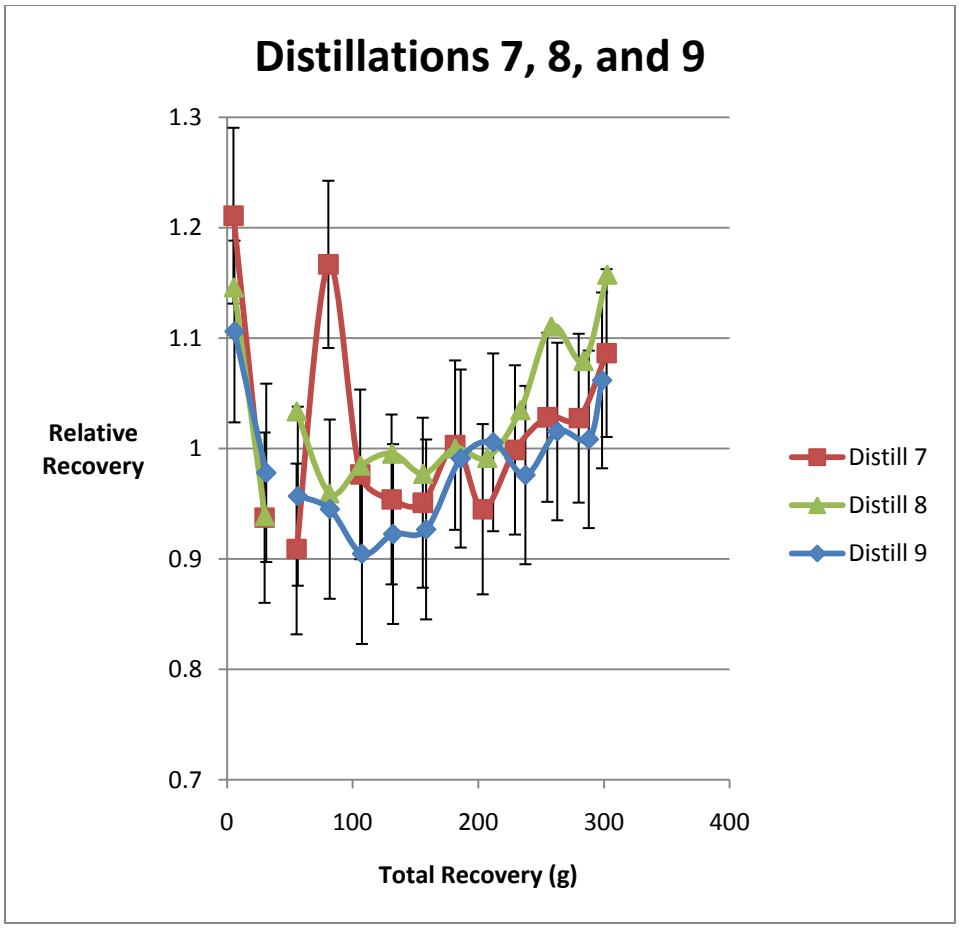

Figure 6.2: Distillations heated on highest heat setting (#10) until after the first drop of distillate, uncertainties are reported at k=2.

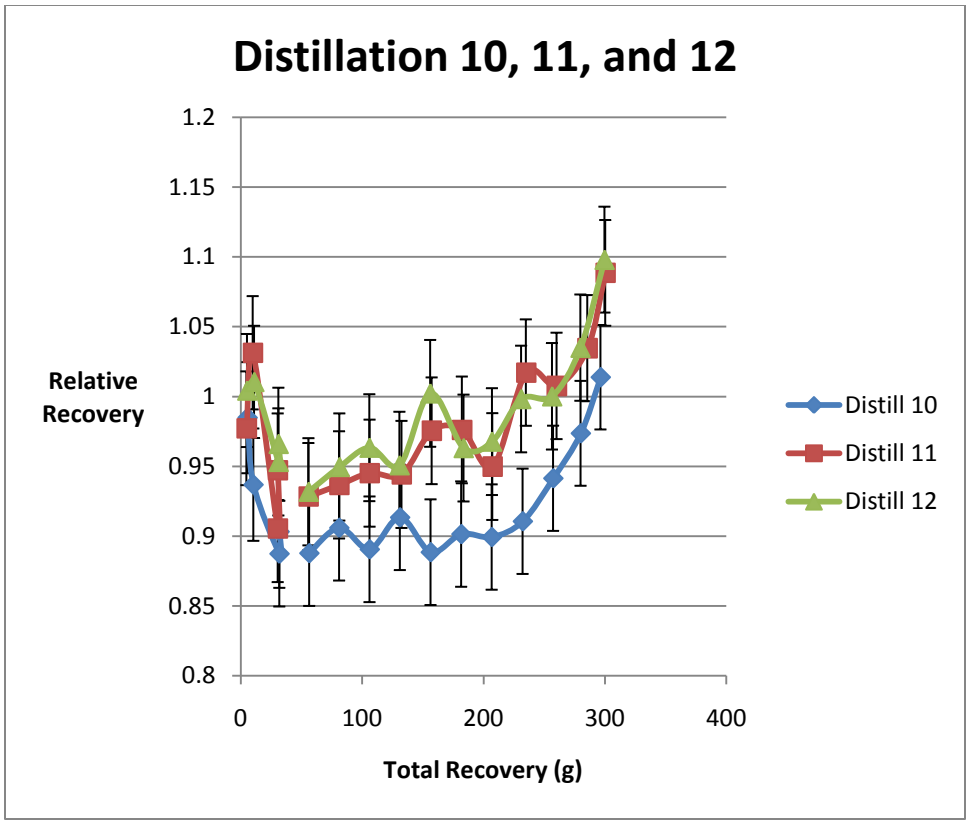

Figure 6.3: Distillations heated on heat setting #8 the entire time, uncertainties are reported at k=2.

The graphs for Distillations 7, 8, and 9 are very similar except for one data point in Distillation 7 that was noticeably higher which appears to be an outlier. However, this outlier does not appear in the first 25 mL, such that the relative recovery result and its uncertainty are not affected.

The heat setting for Distillations 7, 8, and 9 was turned down after the first drops of distillate were collected in the condensate receptacle, causing the average recovery of the first 25 mL to be slightly lower. The step of reducing the temperature is not recommended because this adds another variable to control when developing correction factors.

The average activity concentration recovery decreased in Distillations 10, 11, and 12 when the heat setting was on the lower heat setting the entire time. The relative recoveries of all the

distillations are compared and displayed in Tables 6.3-6.5. Since the activity concentration recovery for the first 25 mL declines as the heat setting is lowered, the CF therefore depends on the heat setting chosen by the operator. These correction factors can only be used for the heat setting for distillations at CSU because of the differences in the distillation setups between SRS and CSU. The t-values below compare the first 25 mL activity concentration to the original activity concentration of the 1000 mL standard. A low t-values indicates the relative activity concentration recovery is essentially 1.

Table 6.3 Isotopic effect in the first 25 mL in Distillations 5 and 6

Distillation Sequence	Activity Concentration of First 25 mL (Bq/g)	Relative Recovery of First 25 mL	Relative Uncertainty of First 25 mL (k=2)	t-value of First 25 mL
5	0.169	0.946	0.070	1.536
6	0.156	0.989	0.091	0.232
	Average	0.968		
	Standard Deviation	0.031		

Table 6.4 Isotopic effect in the first 25 mL in Distillations 7, 8, and 9

Distillation Sequence	Activity Concentration in First 25 mL (Bq/g)	Relative Recovery of First 25 mL	Relative Uncertainty of First 25 mL (k=2)	t-value of First 25 mL
7	0.155	0.937	0.077	1.624
8	0.151	0.939	0.081	1.505
9	0.172	0.978	0.076	0.582
	Average	0.951		
	Standard Deviation	0.022		

Table 6.5 Isotopic effect in the first 25 mL in Distillations 10, 11, and 12

Distillation Sequence	Activity Concentration in First 25 mL (Bq/g)	Relative Recovery of First 25 mL	Relative Uncertainty of First 25 mL (k=2)	t-value of First 25 mL
10	0.674	0.903	0.041	4.720
11	0.673	0.947	0.040	2.635
12	0.679	0.966	0.041	1.655
	Average	0.939		
	Standard Deviation	0.032		

As apparent from the graphs in Figures 6.1-6.3, the activity concentration in the first 5 mL is always substantially higher than for subsequent aliquots. The graphs indicate that a higher heat setting produces a higher relative recovery in the first 5 mL. SRS protocol discards the first 5 mL for this reason. In Distillations 10, 11, and 12, the first 25 mL vials were segmented into a 5 mL and 20 mL vial to obtain more information about the isotopic effect. The relative recoveries of the first 5 mL, second 5 mL, and 20 mL are compared in Table 6.6. The mass recovery at which the isotopic effect stops cannot be determined from the information available. The results might have been more definitive if a higher heat setting had been used due to the increased HTO/H_2O effect in the first 5 mL as demonstrated earlier.

Table 6.6 Comparison of relative recoveries of the first two 5 mL and the first 25 mL.

Distillation Sequence	First 5 mL	Second 5 mL	Change	20 mL	Second 5 + 20 mL
10	0.984846	0.936887	-0.04796	0.88747765	0.903254
11	0.97727	1.031324	0.054055	0.90554254	0.94726
12	1.004295	1.031324	0.027029	0.95317816	0.966087

Since the activity concentration of the first 5 mL was always higher and the activity concentration of the first 25 mL was always lower than expected, small variations in the volume discarded could result in significantly different activity concentration recovery. Discarding exactly 5 mL every time is difficult, requiring an additional mass measurement during the

condensation process. To remove potential sources of variation or bias in discarding the first 5 mL, it may be more accurate and easier not to discard that aliquot. This protocol could save one vial for each distillation and be more efficient in subsequent processes. The total activity concentration recovery can be estimated by combining the first 5 mL and first 25 mL by the weighted average of Distillations 5, 7, 8, and 9. These are compared in Table 6.7 which were chosen for the final CF because they provide the most upper bound for the uncertainties. Distillations 10, 11, and 12 are compared separately in Table 6.8 because of the lower heat setting used during distillation.

Table 6.7 Comparison of relative recoveries for Distillations 5, 7, 8, and 9 if the first 5 mL were combined with the first 25 mL.

Distillation Sequence	5 mL Relative Recovery	25 mL Relative Recovery	Combined 30 mL Relative Recovery
5	1.27322521	0.945963	1.056978
6	1.22548933	0.989387	1.071512
7	1.21079735	0.937394	1.028492
8	1.14610022	0.938929	1.011317
9	1.10595199	0.977961	1.024822
		Average	1.038624

Table 6.8 Comparison of relative recoveries for Distillations 10,11, and 12 if the first 5 mL and second 5 mL were combined with the next 20 mL.

Distillation	5 mL Relative Recovery	25 mL Relative Recovery	Combined 30 mL Relative Recovery
10	0.984846	0.903254	0.925619
11	0.97727	0.94726	0.95444
12	1.004295	0.966087	0.98139
		Average	0.953816

Table 6.7 shows that if boiled more vigorously the relative recovery average is 3.9% higher than expected. Table 6.8 shows that if boiled less vigorously the relative recovery average is 4.6% lower than expected.

The relative recovery uncertainties are calculated according to the following equations. Since the distillation sample was made from the same Eckert & Ziegler standard, the calibration standard uncertainty (+/-20%) does not need to be included in the further considerations. The fractional recovery is given by equation 6.2.

$$CF = \frac{C_{sample-blank}}{C_{1000mlstandard-blank}} \tag{6.1}$$

$$C_{sample-blank} = C_{sample\_measurement} - C_{blank\_measurement} \tag{6.2}$$

$$C_{1000mlstandard-blank} = C_{1000mlstandard\_measurement} - C_{blank\_measurement} \tag{6.3}$$

where $C_{1000mlstandard\_measurement}$ is the activity concentration of the diluted standard, and $C_{1000mlstandard-blank}$ is the activity concentration of the 1000 mL standard minus the activity concentration of the blank.

The relative uncertainty, $\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)$, on the activity concentration of the sample measurement is calculated according to

$$\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2 = \left(\frac{\sigma_{scale}}{m_{scale}}\right)^2 + \left(\frac{\sigma_{instrument}}{C_{instrument}}\right)^2 + \left(\frac{\sigma_{sample-counts}}{n_{sample}}\right)^2 \tag{6.4}$$

where $\left(\frac{\sigma_{scale}}{m_{scale}}\right)$ is the relative uncertainty on the mass determined by the appropriate scale as given in Appendix A, $\left(\frac{\sigma_{instrument}}{C_{instrument}}\right)$ is the relative uncertainty of the CSU LSC machine discussed

in Chapter 4, and $\left(\frac{\sigma_{sample-counts}}{n_{sample}}\right)$ is the relative statistical counting uncertainty of the sample measurement.

The relative uncertainty, $\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)$, on the activity concentration of the blank measurement is calculated according to

$$\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)^2 = \left(\frac{\sigma_{scale}}{m_{scale}}\right)^2 + \left(\frac{\sigma_{instrument}}{C_{instrument}}\right)^2 + \left(\frac{\sigma_{blank\_counts}}{n_{blank}}\right)^2 \quad (6.5)$$

where $\left(\frac{\sigma_{blank\_counts}}{n_{blank}}\right)$ is the relative statistical counting uncertainty on the blank measurement.

The blank is subtracted from the sample activity concentration measurement. Uncertainties of measurements that are added or subtracted must be converted back to absolute values according to:

$$\sigma_{sample-blank} = \sqrt{C^2_{sample\_measurement}\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2 + C^2_{blank\_measurement}\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)^2} \quad (6.6)$$

where the $\sigma_{sample-blank}$ is the uncertainty on the sample measurement when the blank measurement is subtracted.

The relative uncertainty, $\left(\frac{\sigma_{1000ml standard\_measurement}}{C_{1000ml standard\_measurement}}\right)$, on the activity concentration measurement in the 1000 mL standard is calculated according to:

$$\left(\frac{\sigma_{1000ml standard\_measurement}}{C_{1000ml standard\_measurement}}\right)^2 = \left(\frac{\sigma_{scale}}{m_{scale}}\right)^2 + \left(\frac{\sigma_{instrument}}{C_{instrument}}\right)^2 + \left(\frac{\sigma_{1000ml standard\_counts}}{n_{1000ml standard}}\right)^2 \quad (6.7)$$

where $\left(\frac{\sigma_{1000ml standard\_counts}}{n_{1000ml standard}}\right)$ is the relative statistical counting uncertainty on the 1000 mL standard activity concentration measurement.

The blank is subtracted from the 1000 mL standard activity concentration measurement. Uncertainties of measurements that are added or subtracted must be converted back to absolute values according to:

$$\sigma_{1000ml standard-blank} = \sqrt{C^2_{1000ml standard\_measurement}\left(\frac{\sigma_{1000ml standard\_measurement}}{C_{1000ml standard\_measurement}}\right)^2 + C^2_{blank\_measurement}\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)^2} \quad (6.8)$$

where $\sigma_{1000ml standard-blank}$ is the uncertainty on the 1000 mL standard activity concentration measurement when the blank is subtracted.

Since terms appear in the form of a division in equation 6.1, the uncertainties are converted back to relative terms according to:

$$\frac{\sigma_{CF}}{C_{sample\_measurement}} = \sqrt{\left(\frac{\sigma_{sample-blank}}{C_{sample-blank}}\right)^2 + \left(\frac{\sigma_{1000ml standard-blank}}{C_{1000ml standard-blank}}\right)^2} \quad (6.9)$$

Then multiply $\frac{\sigma_{CF}}{C_{sample\_measurement}}$ by a factor of 100 to obtain a result in terms of percent:

$$CF = \frac{C_{sample-blank}}{C_{1000ml standard-blank}} \pm \frac{\sigma_{CF}}{C_{sample\_measurement}}\% \quad (6.10)$$

c) Absorption and Desorption Correction Factor and Uncertainty Estimation

i) 20 mL Vial Absorption

The goal is to determine the absorption, instability, and other unknown effects that result from time delay in counting the 20 mL vials. Previously, the stability of different cocktails over a time period of 60 days was investigated (Barquero and Arcos, 2000). One of the cocktails tested in that study was Ultima Gold, but the particular part number is not provided. The

experiments reported in the literature used glass vials. For comparison, results from the previous study are extracted from a graph in their paper. These approximate values are compared to the results of the current study in Table 6.9. Barquero and Arcos reported approximately a 15% loss of activity over 60 days in glass vials. The maximum loss in glass vials was measured at 2% in this study. Varying biodegradability of the cocktails used by the previous study and this study might account for this difference.

In our study, the same cocktail and concentration were used for both plastic and glass vials. The most notable degradation was for the lower activity concentration. The activity loss in plastic vials for each concentration over 63 days was approximately 16% (0.16 Bq/g), 10% (0.67 Bq/g) , and 0.0% (18.6 Bq/g). Since the glass and the plastic test results were drastically different at lower activity concentrations, cocktail degradation does not explain the activity concentration loss. For Ultima Gold LLT used in this study, the approximate 3.3% degradation in the glass vial over 64 days can be deemed due to cocktail instability. Because this study and previous data were decay corrected, the additional activity concentration loss in the plastic vial may be attributable to absorption.

Table 6.9. Degradation of 20 mL vial activity over time.

	Degradation After Delay (d)						
Experiment	1	3	6	8	38	60	63
Barquero 1999	0.995	0.986	0.978	0.969	0.891	0.851	
0.16 Bq/g plastic	0.892	0.741	0.847	0.804	0.824		0.838
0.16 Bq/g glass	1.034	0.979	0.991	1.023	0.958		0.967
0.67 Bq/g plastic	1.200	0.958	0.883	0.918	0.950		0.900
0.67 Bq/g glass	0.979	0.992	1.011	1.003	0.975		0.987
18.6 Bq/g plastic	1.005	1.005	1.004	1.007	1.008		1.004
18.6 Bq/g glass	1.001	0.999	0.999	0.999	1.000		1.002

An activity concentration reduction correction factor was calculated by graphing the data in Appendix D versus time in Figures 6.4-6.9; an exponential trend line is used to find an analytical equation for this correction factor. The exponential trend line was chosen because

diffusion is usually described by a first order kinetics, which, upon integration, results in an exponential expression for the time dependency. In order to verify our results, the data to be modified by the activity concentration reduction factor in the later experiments are graphed in Figures 6.10-6.12. The slopes of those trend lines in the later section are consistent with the slopes of the Figures 6.4-6.9 which seem to support the validity of equations 6.11- 6.13.

The error bars in the graphs represent the combined counting, scale, and instrument uncertainties (k=2). All graphs follow a similar slope except the standard in the glass vial with an activity concentration of 18.6 Bq/g. The increasing trend of the glass vial with approximate activity concentration of 18.6 Bq/g is not significant since the t-value in Appendix D is below 1.5 as chosen for our study indicating there was no absorption or desorption. According to data from this study, no correction factor would be needed for glass vials or plastic vials with an activity concentration of 18.6 Bq/g.

One data point was omitted in the graph displaying the data for the 20 mL plastic vials with an activity concentration of 0.67 Bq /g. The activity concentration appears to reach 182% of the initial concentration which is not physically possible and significantly altered the analytical fit parameters. Omitting this outlier resulted in a similar slope to the three other similar concentrations in Figures 6.4, 6.10, and 6.11.

The instrument uncertainty for vials with an activity concentration of 0.37 Bq/g was not established independently. The instrument uncertainty for that particular activity concentration was derived by averaging the uncertainty values obtained from vials containing activity concentrations of 0.16 and 0.67 Bq/g .

The analytical fit equations for the activity concentration reduction equations are given by:

$CF_{0.16Bq/g\ counting\ delay} = 0.08763e^{-0.002t}$, t is in days (6.11)

for vials containing an approximate activity concentration of 0.16 Bq/g,

$CF_{0.67Bq/g\ counting\ delay} = 1.0000e^{-0.002t}$, t is in days (6.12)

for vials containing an approximate activity concentration of 0.67 Bq/g, and

$CF_{18.6Bq/g\ counting\ delay} = 1.0002e^{-0.002t}$, t is in days (6.13)

for vials containing an approximate activity concentration of 18.6 Bq/g.

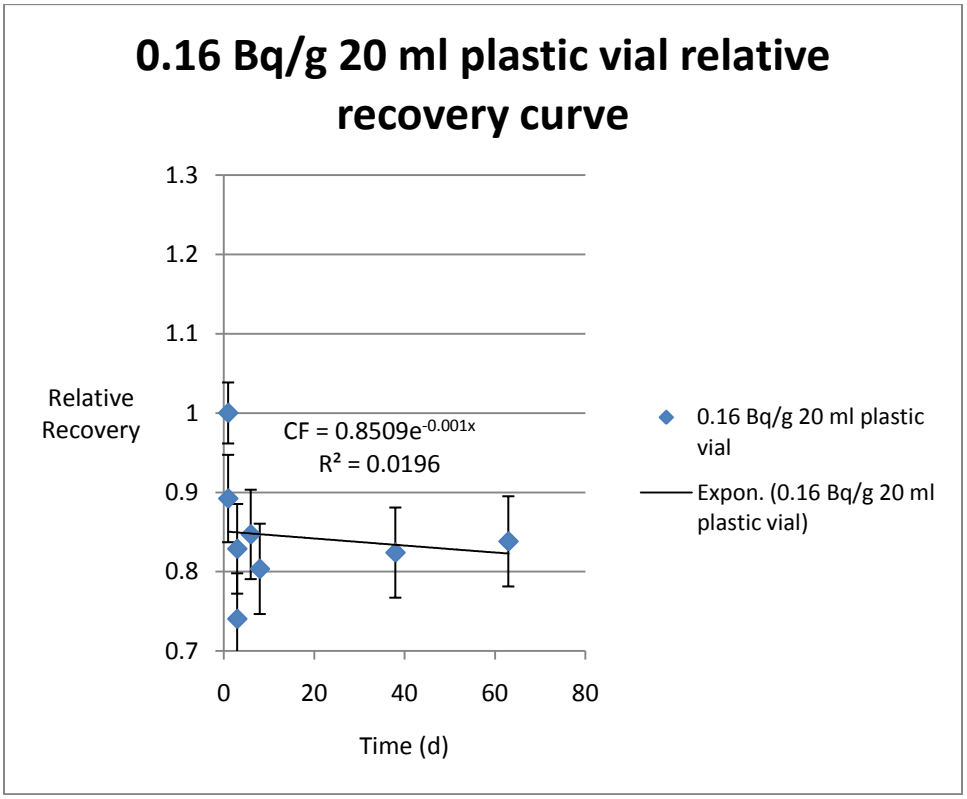

Figure 6.4. 20 mL plastic vial with an activity concentration of 0.16 Bq/g, relative recovery curve, uncertainties are reported at k=2.

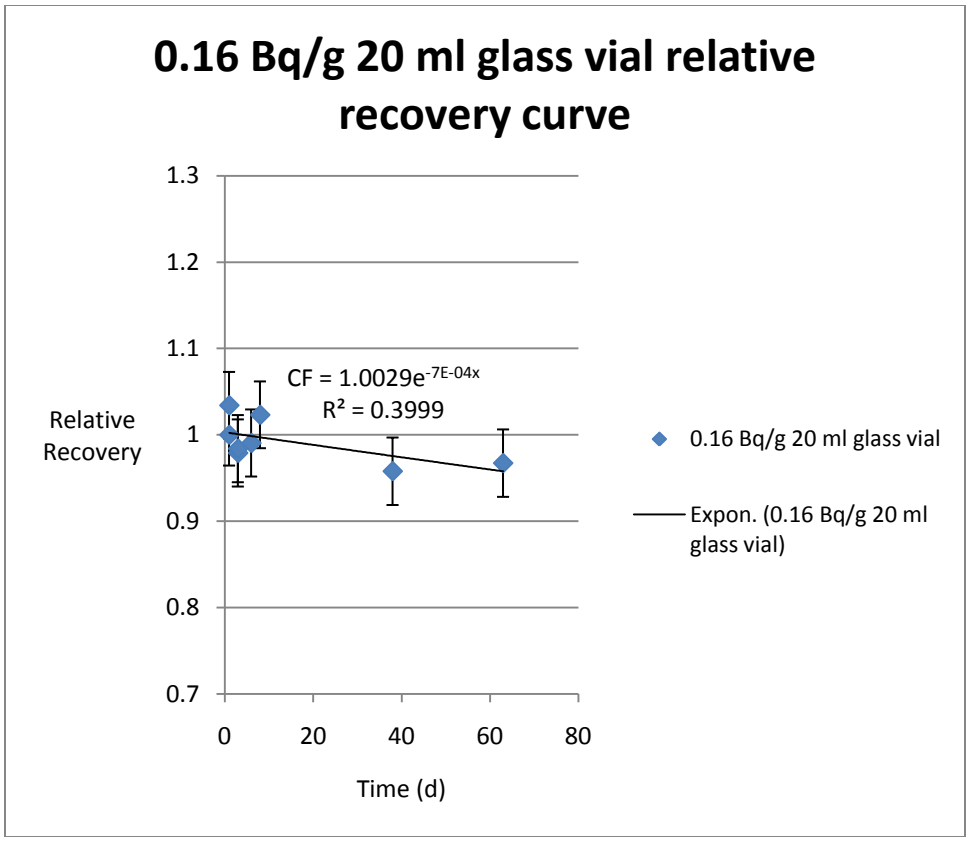

Figure 6.5. 20 mL glass vial with an activity concentration of 0.16 Bq/g, relative recovery curve, uncertainties are reported at k=2.

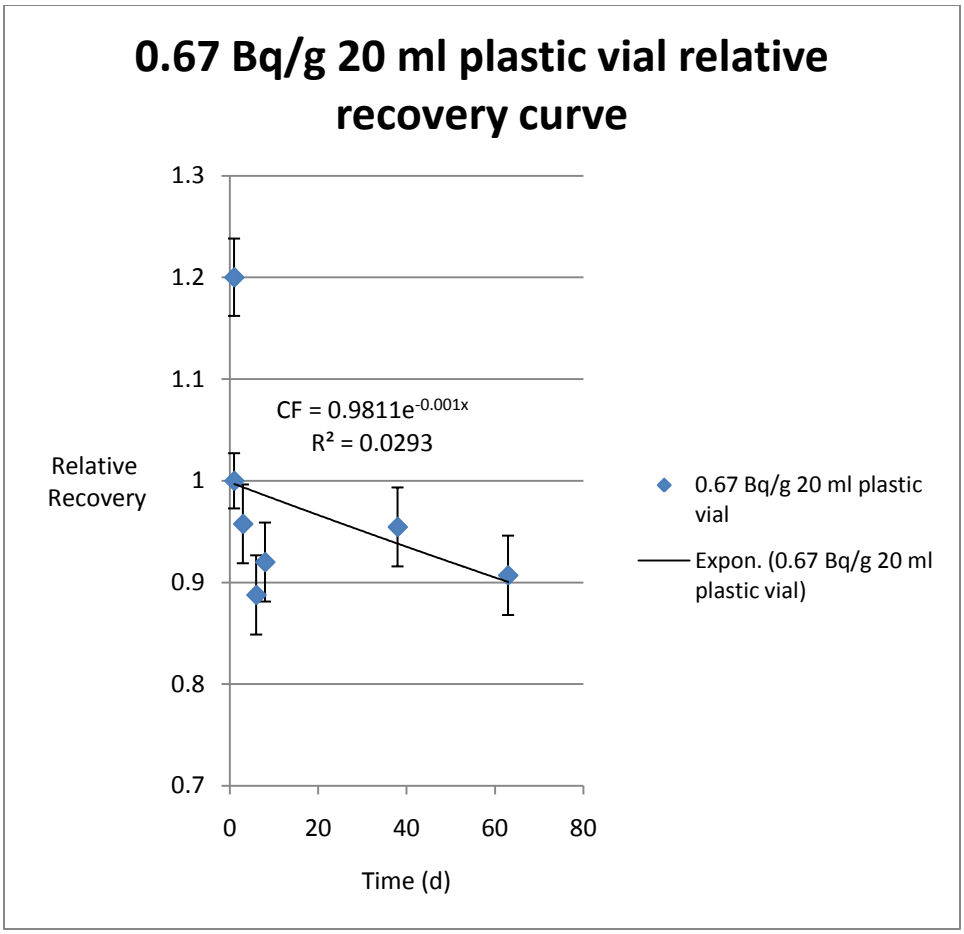

Figure 6.6. 20 mL plastic vial with an activity concentration of 0.67 Bq/g, relative recovery curve, uncertainties are reported at k=2.

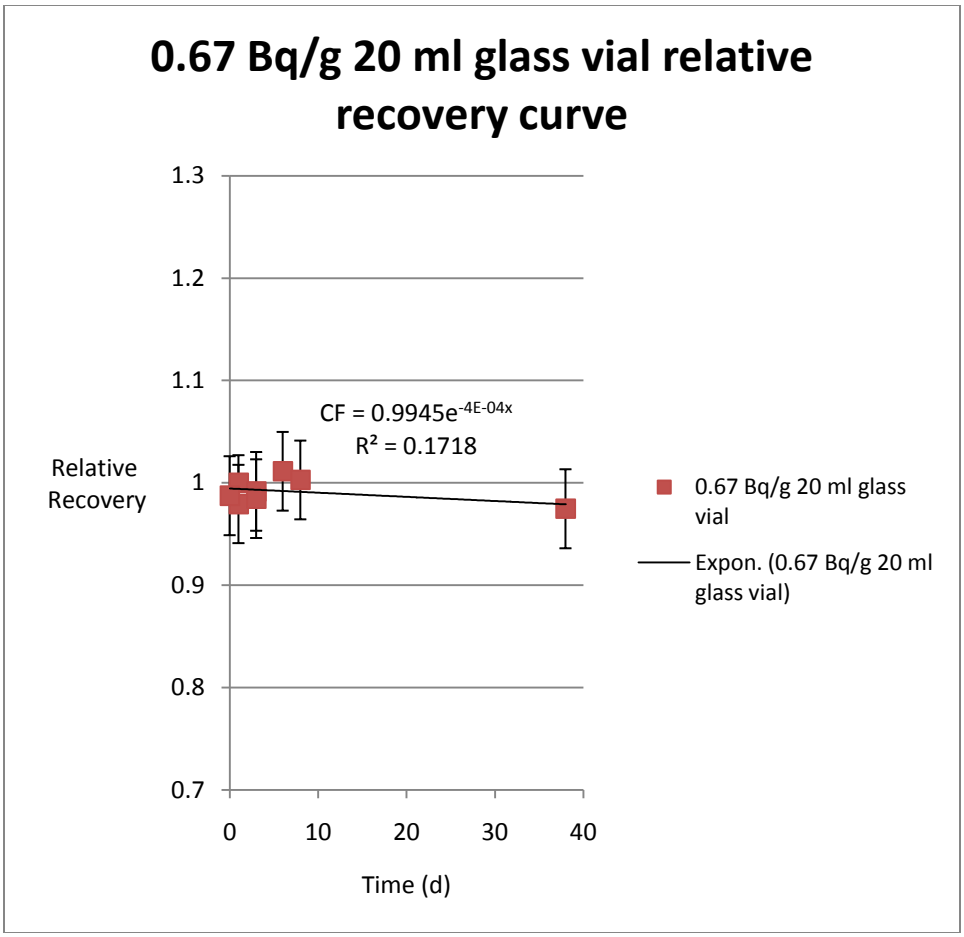

Figure 6.7. 20 mL glass vial with an activity concentration of 0.67 Bq/g, relative recovery curve, uncertainties are reported at k=2.

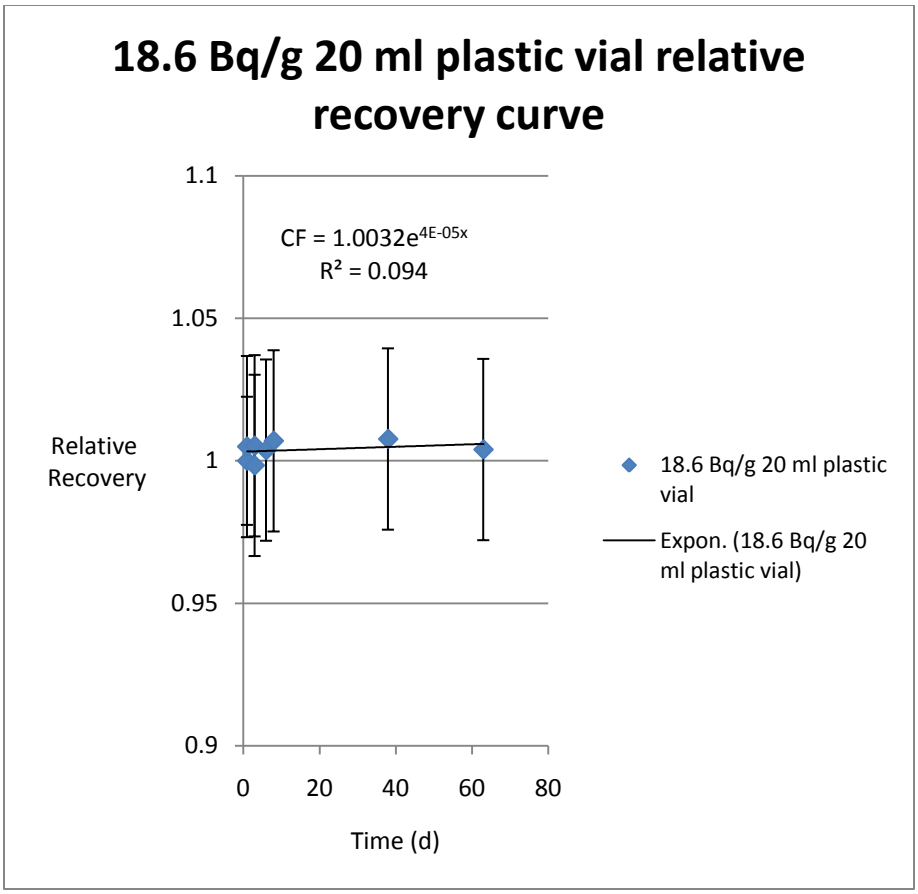

Figure 6.8. 20 mL plastic vial with an activity concentration of 18.6 Bq/g, relative recovery curve, uncertainties are reported at k=2.

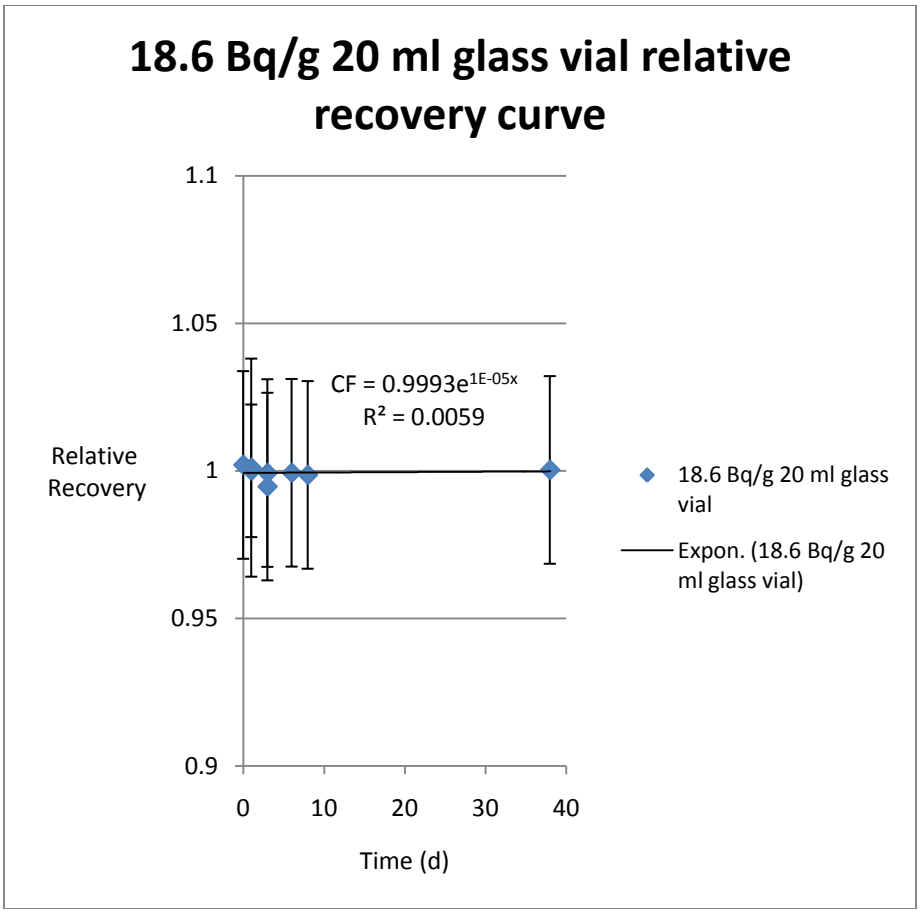

Figure 6.9. 20 mL glass vial with an activity concentration of 18.6 Bq/g, relative recovery curve, uncertainties are reported at k=2.

The error bars in Figures 6.4 - 6.9 and the corresponding tables are calculated according to the following equations. The activity concentration recovery equation is given by:

$$C_{Recovery}(t) = \frac{C_{sample-blank}(t)}{C_{initial-blank}(t)} \tag{6.14}$$

$$C_{sample-blank} = C_{sample\_measurement} - C_{blank\_measurement} \tag{6.15}$$

$$C_{initial-blank} = C_{initial\_measurement} - C_{blank\_measurement} \tag{6.16}$$

where $C_{initial\_measurement}$ is the activity concentration of the first sample, and $C_{initial-blank}$ is the activity concentration of the first sample minus the activity concentration of the blank.

The calculation of the uncertainty on the recovery follows the methodology outlined in Chapter 2. Specifically, the following equations have been derived for the uncertainty on the activity concentration recovery fraction:

$$\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2 = \left(\frac{\sigma_{scale}}{m_{scale}}\right)^2 + \left(\frac{\sigma_{instrument}}{C_{instrument}}\right)^2 + \left(\frac{\sigma_{sample\_counts}}{n_{sample}}\right)^2 \tag{6.17}$$

$$\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)^2 = \left(\frac{\sigma_{scale}}{m_{scale}}\right)^2 + \left(\frac{\sigma_{instrument}}{C_{instrument}}\right)^2 + \left(\frac{\sigma_{blank\_counts}}{C_{blank}}\right)^2 \tag{6.18}$$

The blank is subtracted from the sample concentration measurement. Uncertainties of measurements that are added or subtracted must be converted backed to absolute values according to:

$$\sigma_{sample-blank} = \sqrt{C^2_{sample\_measurement}\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2 + C^2_{blank\_measurement}\left(\frac{\sigma_{blank\_measurement}}{C_{blank\_measurement}}\right)^2} \tag{6.19}$$

Where $\sigma_{sample-blank}$ is the relative uncertainty of the sample measurement when the blank is subtracted.

The relative uncertainty, $\left(\frac{\sigma_{initial-blank}}{C_{initial-blank}}\right)$, on the initial sample activity concentration is the uncertainty on the first sample and calculated in analogy to equation 6.19.

Since the individual terms in equation 6.14 appear in a form of division, the terms are converted back to a relative term according to:

$$\frac{\sigma_{recovery}}{C_{sample\_measurement}} = \sqrt{\left(\frac{\sigma_{sample-blank}}{C_{sample-blank}}\right)^2 + \left(\frac{\sigma_{initial-blank}}{C_{initial-blank}}\right)^2} \tag{6.20}$$

When $\left(\frac{\sigma_{recovery}}{C_{sample\_measurement}} \right)$ is multiplied by a factor of 100 the resulting relative uncertainty is expressed in terms of percent.

$$C_{recovery}(t) = \frac{C_{sample-blank}(t)}{C_{initial-blank}} \pm \frac{\sigma_{recovery}}{C_{sample\_measurement}} \% \tag{6.21}$$

ii) 1000 mL Bottle Absorption

The 1000 mL bottle experiments were designed to identify if any counts were lost due to absorption or other means such as plating while the composite sample is collected for 2 weeks. The experimental data are presented in Appendix E. Measurement data are reported with the activity concentration correction derived in the previous section. The 20 mL vial correction factors extracted from the fit equation in the appropriate figures (6.4, 6.6, and 6.8) greatly improved the interpretation of the data when counting was delayed for a significant amount of time.

The same degradation was observed in the 1000 mL bottle experiments as in the 20 mL plastic vial absorption investigation. The results for the 1000 mL bottle with an activity concentration of approximately 0.16 Bq/g are graphed and an exponential fit to the data is calculated as shown in Figures 6.10 through 6.12. The slopes in Figures 6.10 through 6.12 are consistent with the slope obtained in Figure 6.4. Since this observed effect is comparable to the effect also observed in the 20 mL plastic vial absorption test, the effect can be attributed to the time delay in counting. The measured data from the tests were corrected with the analytic equations for the appropriate activity concentration obtained from the 20 mL vial test of 63 days. The corrected data are consistent with expectation. For comparison, both corrected and uncorrected data are shown in Appendix E.

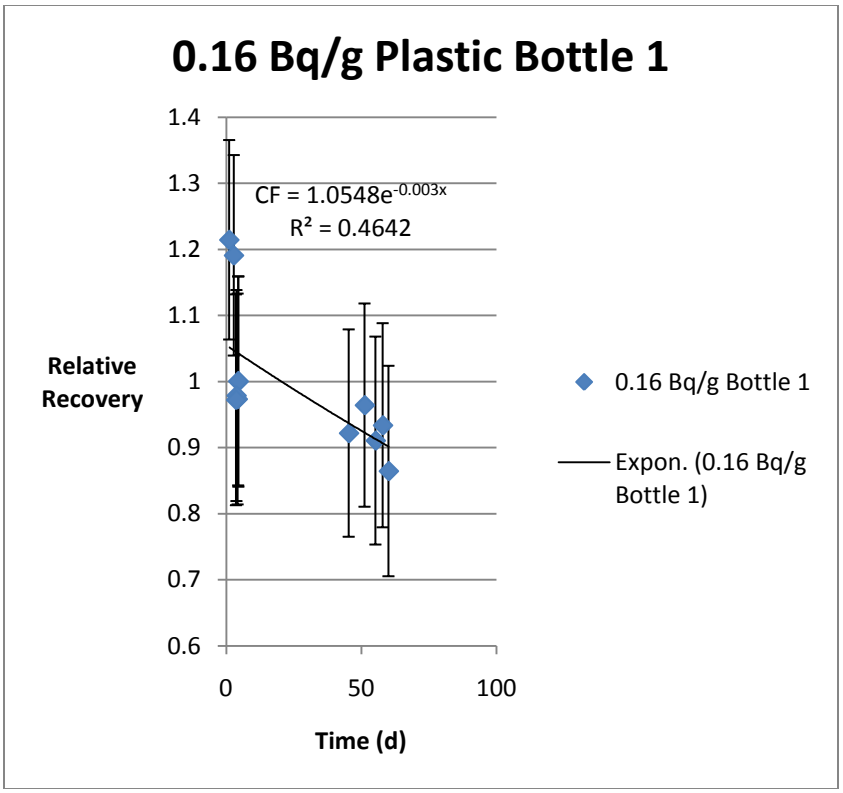

Figure 6.10. 1.0 L Plastic Bottle 1 with an activity concentration of 0.16 Bq/g absorption test, uncertainties are reported at k=2.

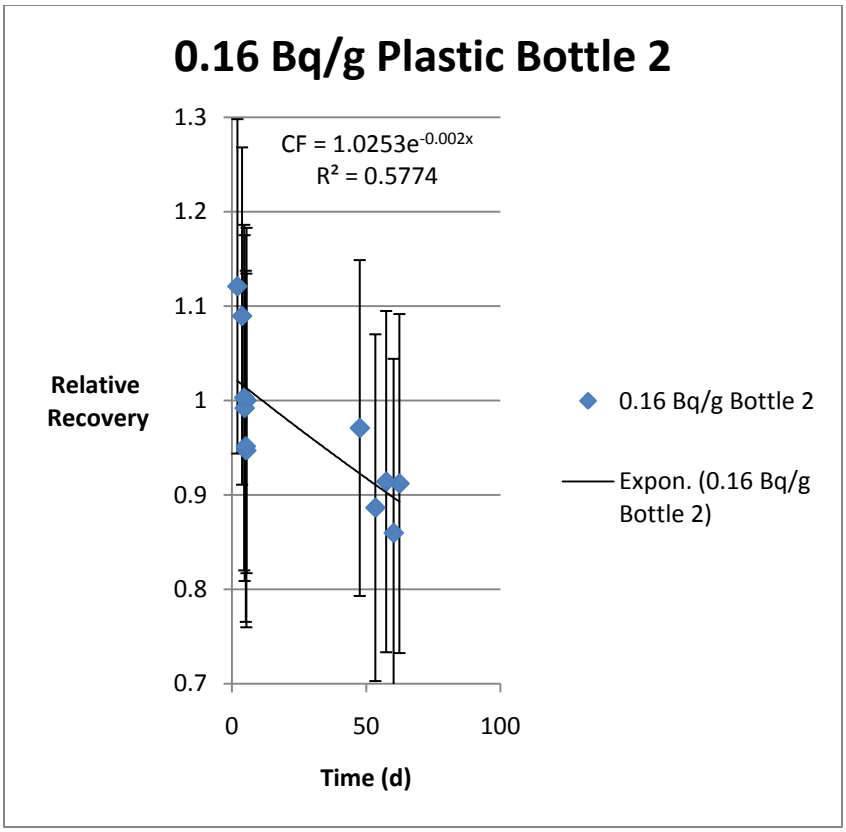

Figure 6.11. 1.0 L Plastic Bottle 2 with an activity concentration of 0.16 Bq/g absorption test, uncertainties are reported at k=2.

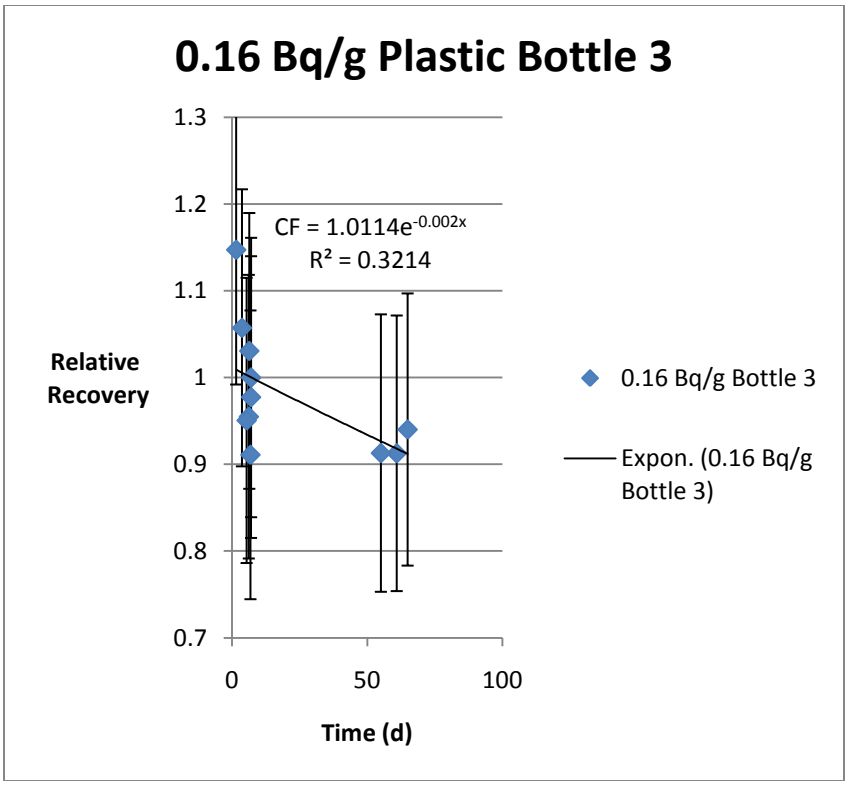

Figure 6.12. 1.0 L Plastic Bottle 3 with an activity concentration of 0.16 Bq/g absorption test, uncertainties are reported at k=2.

The individual fractional recovery data points in each of the experiments displayed in Appendix E fluctuate about 1.0 or display positive values except for bottles 8 and 9. Bottle 9 is fluctuating about 1.0 except for the last three samples which are declining at the end. The t-value for Bottle 8 is below the established criteria (t=1.5). The result is not statistically significant. Only the last data point for Bottle 9 is statically significant to indicate some sort of absorption. Since the bottles 7 and 8 do not show any absorption, the last data point for bottle 9 may be an outlier.

The initial and final activity concentration reduction corrected data is compared in Tables 6.10 through 6.12. The bottles with activity concentrations of 0.16 Bq/g and 0.37 Bq/g exhibit relative recoveries greater than 1.0 which has to be attributed to the total uncertainty of the measurement. There is no noticeable statistically significant absorption except for bottle 9.

Since two out of the three bottles with activity concentrations of 0.67 Bq/g show absorption, the results are inconclusive. The fractional recovery fluctuations about 1.0 in Appendix E indicate that the observed effect is most likely due to the instrument uncertainty. Also, the 20 mL vial activity concentration reduction trend line downward slope is large and likely masks the possible absorption in 1000 mL bottles.

Table 6.10. Bottles with activity concentrations of 0.16 Bq/g absorption test

Test	Days between Initial and Final	Initial Activity Concentration (Bq/g)	Relative Standard Deviation (k=1)	Final Corrected Activity Concentration (Bq/g)	Relative Standard Deviation (k=1)	Relative Recovery	t-value
Bottle 1	19.3	0.142	0.080	0.142	0.078	1.00	0.003
Bottle 2	19.3	0.142	0.091	0.150	0.089	1.06	0.442
Bottle 3	19.2	0.137	0.081	0.138	0.080	1.00	0.044

Table 6.11. Bottles with activity concentrations of 0.37 Bq/g absorption test

Test	Days between Initial and Final	Initial Activity Concentration (Bq/g)	Relative Standard Deviation (k=1)	Final Corrected Activity Concentration (Bq/g)	Relative Standard Deviation (k=1)	Relative Recovery	t-value
Bottle 4	19.2	0.300	0.043	0.334	0.041	1.12	1.596
Bottle 5	19.1	0.299	0.043	0.337	0.041	1.13	1.796
Bottle 6	19.2	0.306	0.043	0.340	0.041	1.11	1.348

Table 6.12. Bottles with activity concentrations of 0.67 Bq/g absorption test

Test	Days between Initial and Final	Initial Activity Concentration (Bq/g)	Relative Standard Deviation (k=1)	Final Corrected Activity Concentration (Bq/g)	Relative Standard Deviation (k=1)	Relative Recovery	t-value
Bottle 7	19.1	0.513	0.031	0.514	0.032	1.00	0.013
Bottle 8	19.1	0.645	0.023	0.633	0.023	0.98	0.540
Bottle 9	19.1	0.671	0.023	0.626	0.023	0.93	2.089

The error bars in Figures 6.10 through 6.12 and the uncertainties in Tables 6.10 through 6.12 are calculated according to equations 6.17 through 6.20. The recovery is calculated by equations 6.14 through 6.16.

iii) 1000 mL Bottle Desorption

The data for the 1000 mL desorption bottle test are displayed in Appendix F. The first one week desorption test seems to indicate tritium was desorbed with activity concentrations after one week ranging from 103 to 153% above background. Most t-values were above the established criteria (1.5), confirming tritium desorption from the plastic back into the DI water. The average desorption for the bottles with activity concentrations of 0.16 Bq/g, 0.37 Bq/g, and 0.67 Bq/g was 140%, 121%, and 108%, respectively. The bottle 8 test had the lowest desorption at a value of 100.3% with a t-value of 0.589 which means there was no desorption. Eight out of nine results suggest that some tritium did absorb and was released. The bottles that had the highest original concentrations had the lowest desorption which is inconsistent with expectation. The higher concentrations should have had more or at least the same absorption and thus more or the same desorption.

Using the t-value greater than 1.5 criteria, only three out of nine bottles indicate desorption for the second week. The desorption was expected to be less since the bottles had been rinsed twice and the desorption of the first week test removes some of the tritium that could not be removed during the second week. These test results are inconclusive.

The blank and the initial sample are the same in this experiment. The blank can therefore not be subtracted from the initial sample like in the other experiments. The fractional recovery is given by equation 6.22

$$C_{recovery}(t) = \frac{C_{sample}(t)}{C_{DIwater}} \tag{6.22}$$

where $C_{DIwater}$ is the activity concentration of the deionized water used to fill the bottles.

The relative recovery uncertainties displayed in Appendix F are calculated according to the following equations and are displayed at k=2.

$$\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2 = \left(\frac{\sigma_{scale}}{m_{scale}}\right)^2 + \left(\frac{\sigma_{instrument}}{C_{instrument}}\right)^2 + \left(\frac{\sigma_{sample\_counts}}{n_{sample}}\right)^2 \quad (6.23)$$

$$\left(\frac{\sigma_{DIwater}}{C_{DIwater}}\right)^2 = \left(\frac{\sigma_{scale}}{m_{scale}}\right)^2 + \left(\frac{\sigma_{instrument}}{C_{instrument}}\right)^2 + \left(\frac{\sigma_{DIwater\_counts}}{n_{DIwater}}\right)^2 \quad (6.24)$$

where $\sigma_{DIwater}$ is the uncertainty on the activity concentration of the deionized water, $C_{DIwater}$ is the activity concentration of the deionized water, and $\left(\frac{\sigma_{DIwater\_counts}}{n_{DIwater}}\right)$ is the relative statistical counting uncertainty of the deionized water measurement.

The relative uncertainty on the recovery is calculated according to:

$$\frac{\sigma_{recovery}}{C_{sample\_measurement}} = \sqrt{\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2 + \left(\frac{\sigma_{DIwater}}{C_{DIwater}}\right)^2} \quad (6.25)$$

$\frac{\sigma_{recovery}}{C_{sample\_measurement}}$ is multiplied by a factor of 100 to yield a result in terms of percent.

$$C_{Recovery}(t) = \frac{C_{sample}(t)}{C_{DIwater}} \pm \frac{\sigma_{recovery}}{C_{sample\_measurement}}\% \quad (6.26)$$

Chapter 7 Conclusions

Since the first 5mL was always higher than the original concentration in the distillation experiments, correction factors for the HTO/H_2O isotopic effect would have to be calculated for different discarded volumes. This would require an extra step of measuring the first 5 mL volume. There is a possibility to simplify the distillation process by not discarding the first 5 mL. It would remove the variability in measuring the volume of the discard, potentially impacting the correction factor calculation, and it would reduce the waste by one 30 mL glass vial per sample distillation. Because the protocol requires discarding the first 5 mL, different correction factors for distillations with different volume discards might have to be established. The point where the relative activity concentration reduction changes from positive to negative values is near the first 5 mL of distillate. Small deviations in the volume discarded may therefore result in large changes in the CF. Additional work to determine the exact volume for where this effect occurs is recommended. The discard volume could be altered to a volume that is predictable or even omitted. The correction factor could become less than zero below a certain heat rate. Table 6.7 shows that if boiled more vigorously the relative recovery average was approximately 3.8% higher than the original concentration. Table 6.8 shows that if boiled less vigorously the relative recovery average was approximately 95% of what it should be. Most regulations usually require a licensee to apply the most conservative value (higher number). Regulators and the public might be adverse to a reduction in the SRS reported measurements. The CF needs to be determined for the appropriate heating rate.

An estimated total CF is calculated according to equation 2.9. The estimated $CF_{pipette}$ is 0.998 as discussed in Chapter 4. The estimated $CF_{HTO\_Isotopic\_Effect}$ is the average correction factor from Distillations 5 - 9 with a value of 1.044. $CF_{Absorption}$ was set to 1 since no absorption was

found in the 1000 mL bottle absorption experiments discussed in Chapter 6. The $CF_{Desorption}$ was set to 1 because the tests discussed in Chapter 6 used activity concentrations significantly higher than any recent measured activity concentration in the Savannah River. The estimated total CF is $CF = 0.998 \times 1.044 \times 1 \times 1 = 1.042$.

An estimated uncertainty on the activity concentration in the sample is calculated according to equation 2.16 if the uncertainty on the blank measurements is neglected. The estimated $\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}} \right)$ is calculated according to:

$$\left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}} \right)^2 = (0.0029)^2 + \left(\frac{\sigma_{SRS\_sample\_counts}}{n_{sample}} \right)^2$$

where $\left(\frac{\sigma_{SRS\_sample\_counts}}{n_{sample}} \right)$ is the relative counting uncertainty on the SRS sample activity concentration.

The estimated $\left(\frac{\sigma_{standard\_measurement}}{C_{standard\_measurement}} \right)$ is calculated according to equation 2.14. The number of counts is unknown. Inserting the known pipette calibration value yields

$$\left(\frac{\sigma_{standard\_measurement}}{C_{standard\_measurement}} \right)^2 = (0.0029)^2 + \left(\frac{\sigma_{SRS\_standard\_counts}}{n_{standard}} \right)^2$$

where $\left(\frac{\sigma_{SRS\_standard\_counts}}{n_{standard}} \right)$ is the relative counting uncertainty on the SRS standard activity concentration.

The average HTO/H_2O isotopic effect uncertainty for Distillations 5-9 was 0.08 at k = 2. Since the SRS calibration standard uncertainty is unknown, the CSU calibration standard uncertainty (+/- 20%, k=2) will be used.

The estimated total relative uncertainty $\left(\frac{\sigma_{total}}{C_{sample\_measurement}}\right)$ on the activity concentration in the sample is

$$\frac{\sigma_{total}}{C_{sample\_measurement}} = \sqrt{\left(\frac{\sigma_{standard\_measurement}}{C_{standard\_measurement}}\right)^2 + (0.2)^2 + (0.08)^2 + \left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2}$$

$$\frac{\sigma_{total}}{C_{sample\_measurement}} = \sqrt{0.0464 + \left(\frac{\sigma_{standard\_measurement}}{C_{standard\_measurement}}\right)^2 + \left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2}$$

This value is multiplied by a factor of 100 to express the results in terms of percent. Inputting values into Equation 2.10 then provides an estimated weekly activity concentration sample equation:

$$C_{sample\_reported} = \frac{C_{sample\_measurement} \times 1.042}{C_{standard\_measurement}} C_{standard} \pm 100 \times \sqrt{0.0464 + \left(\frac{\sigma_{standard\_measurement}}{C_{standard\_measurement}}\right)^2 + \left(\frac{\sigma_{sample\_measurement}}{C_{sample\_measurement}}\right)^2}\ \% \quad (7.1)$$

Chapter 8 Future Work

Because of the difference in burners and possible altitude effects, HTO/H_2O isotopic effect correction factors should be developed on SRS equipment at SRS. Consideration should be given to not discarding the first 5 mL of the distilled sample. The 1000 mL absorption test could be repeated with glass vials or with shorter delays in counting to isolate the 20 mL activity concentration degradation effect that might have obscured any absorption results in this study. Also, the use of LDPE rather than HDPE would better simulate the 15 L sample container.

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Appendix A: Colorado State University ERHS FisherScientific Scale CF and uncertainty

FisherScientific A-160 serial # 19983 was calibrated by Sercom on December 30, 2011. Calibration weight mass and uncertainty are neglected because it is orders of magnitude smaller than the calibration weight measurement. The relative correction factor is 5.0005/5 which equals 1.0001. The uncertainty is the standard deviation of nine measurements of the same vial in Table D.3.

Table A.1. Calibration Weights Information

Nominal Weight (g)	True Mass (g)	Uncertainty (+/-mg)
5	5.000025	0.0073
100	100.0001	0.044

Table A.2. Calibration Weight Measurement on 12/30/2011

Nominal Weight (g)	True Mass (g)
5	5.0005

Table A.3. ERHS Fisher Scientific Scale Uncertainty Determination Using Approximately 10 mL of DI Water and 12 mL of Ultima Gold LLT

Trial #	Weight (g)
1	19.7772
2	19.7421
3	19.7621
4	19.8798
5	19.7665
6	19.8268
7	19.7804
8	19.9047
9	19.8465
Mean	19.80957
Standard Deviation	0.05723
Relative Standard Deviation	0.002889

Appendix B: LSC Instrument Uncertainty Data

Table B.1: LSC Instrument Uncertainty Data for the Plastic Vials with the Approximate Activity Concentration of 0.16 Bq/g

Plastic Vial	Counts	Activity Concentration (Bq/g)	Relative Instrument Counting Uncertainty (k=1)
1	5597	0.220	0.013
2	5405	0.221	0.014
3	5232	0.210	0.014
4	5480	0.219	0.014
5	5410	0.211	0.014
6	5386	0.218	0.014
	Average Measurement (Bq/g)	0.217	
	Relative Deviation (k=1)	0.022	

Note: Optimized ROI was channels 60 to 171

Table B.2: LSC Instrument Uncertainty Data for the Glass Vials with the Approximate Activity Concentration of 0.16 Bq/g

Glass Vial	Counts	Activity Concentration (Bq/g)	Relative Instrument Counting Uncertainty (k=1)
1	7134	0.301	0.012
2	7159	0.301	0.012
3	7157	0.305	0.012
4	7125	0.299	0.012
5	7101	0.292	0.012
6	7097	0.303	0.012
	Average Measurement (Bq/g)	0.300	
	Relative Deviation (k=1)	0.015	

Note: Optimized ROI was channels 60 to 171

Table B.3: LSC Instrument Uncertainty Data for the Plastic Vials with the Approximate Activity Concentration of 0.67 Bq/g

Plastic Vial	Counts	Activity Concentration (Bq/g)	Relative Instrument Counting Uncertainty (k=1)
1	19142	0.731	0.007
2	18985	0.722	0.007
3	18909	0.735	0.007
4	19002	0.722	0.007
5	19044	0.735	0.007
6	18538	0.716	0.007
	Average Measurement (Bq/g)	0.727	
	Relative Deviation (k=1)	0.011	

Note: Optimized ROI was channels 60 to 171

Table B.4: LSC Instrument Uncertainty Data for the Glass Vials with the Approximate Activity Concentration of 0.67 Bq/g

Glass Vial	Counts	Activity Concentration (Bq/g)	Relative Instrument Counting Uncertainty (k=1)
1	20840	0.791	0.007
2	20793	0.787	0.007
3	20864	0.797	0.007
4	20472	0.778	0.007
5	20781	0.810	0.007
6	20613	0.794	0.007
	Average Measurement (Bq/g)	0.793	
	Relative deviation (k=1)	0.014	

Note: Optimized ROI was channels 60 to 171

Table B.5: LSC Instrument Uncertainty Data for the Plastic Vials with the Approximate Activity Concentration of 18.6 Bq/g

Plastic Vial	Counts	Activity Concentration (Bq/g)	Relative Instrument Counting Uncertainty (k=1)
1	54084.61	18.028	0.004
2	54206.77	18.069	0.004
3	54256.31	18.085	0.004
4	54326.98	18.109	0.004
5	54378.99	18.126	0.004
6	54400.33	18.133	0.004
	Average Measurement (Bq/g)	18.092	
	Relative deviation (k=1)	0.002	

Note: Optimized ROI was channels 60 to 171

Table B.6: LSC Instrument Uncertainty Data for the Glass Vials with the Approximately Concentration of 18.6 Bq/g

Glass Vial	Counts	Activity Concentration (Bq/g)	Relative Instrument Counting Uncertainty (k=1)
1	499702	17.996	0.001
2	499898	18.079	0.001
3	500664	18.096	0.001
4	500564	18.099	0.001
5	501938	18.246	0.001
6	500685	18.134	0.001
	Average Measurement (Bq/g)	18.109	
	Relative deviation (k=1)	0.005	

Note: Optimized ROI was channels 60 to 171

Appendix C: Distillation Data

Table C.1: Distillation 5 Heated on 10 for Entire Distillation

Vial	Sample (g)	Activity Concentration (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
1	5.33	0.265	1.199	0.082	2.007
2	24.83	0.210	0.952	0.082	1.655
3	25.6	0.216	0.986	0.082	0.683
4	25.67	0.218	0.986	0.082	0.597
5	25.62	0.222	1.004	0.081	0.158
6	25.42	0.216	0.981	0.081	0.705
7	25.7	0.226	1.026	0.081	0.860
8	25.92	0.217	0.983	0.081	0.577
9	25.45	0.219	0.994	0.081	0.231
10	25.81	0.229	1.037	0.081	1.232
11	24.8	0.227	1.030	0.081	1.015
12	25.61	0.226	1.024	0.081	0.937
13	6.75	0.260	1.179	0.080	4.274
300 mL std		0.220		0.057	

Note: Optimized ROI was channels 62 to 171
Note: Total 300 mL gram recovery was 0.977

Table C.2: Distillation 6 Heated on 10 for Entire Distillation

Vial	Sample (g)	Activity Concentration (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
1	5.64	0.194	1.053	0.087	1.221
2	24.8	0.156	0.850	0.087	3.430
3	24.44	0.156	0.851	0.087	3.416
4	24.12	0.154	0.836	0.088	3.747
5	25.17	0.149	0.810	0.088	4.291
6	24.88	0.160	0.871	0.087	2.968
7	24.07	0.169	0.921	0.086	1.840
8	24.09	0.155	0.843	0.088	3.589
9	24.29	0.161	0.878	0.087	2.820
10	24.55	0.159	0.865	0.087	3.105
11	24.62	0.156	0.850	0.087	3.439
12	24.28	0.163	0.888	0.086	2.588
13	24.55	0.169	0.921	0.086	1.847
300 mL std		0.184		0.059	

Note: Optimized ROI was channels 62 to 171
Note: Total 300 mL gram recovery was 0.990

Table C.3: Distillation 7 Heated on 10 Until after First Vial

Vial	Sample (g)	Activity Concentration (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
1	5.19	0.200	1.211	0.080	5.295
2	24.8	0.155	0.937	0.077	1.624
3	25.46	0.150	0.909	0.077	2.354
4	25.26	0.193	1.167	0.076	4.405
5	25.32	0.161	0.977	0.077	0.610
6	24.98	0.158	0.954	0.077	1.199
7	24.85	0.157	0.951	0.077	1.275
8	25.72	0.166	1.003	0.077	0.080
9	21.95	0.156	0.945	0.077	1.426
10	25.67	0.165	0.999	0.077	0.033
11	25.77	0.170	1.028	0.077	0.739
12	24.95	0.170	1.027	0.076	0.718
13	22.18	0.180	1.086	0.076	2.275
300 mL std		0.165		0.054	

Note: Optimized ROI was channels 62 to 171
Note: Total 300 mL gram recovery was 0.992

Table C.4: Distillation 8 Heated on 10 Until after First Vial

Vial	Sample (g)	Activity Concentration (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
1	5.58	0.184	1.146	0.082	3.544
2	24.65	0.151	0.939	0.081	1.505
3	25.38	0.166	1.034	0.080	0.844
4	25.84	0.154	0.960	0.081	0.999
5	24.94	0.158	0.985	0.081	0.379
6	25.15	0.160	0.995	0.081	0.112
7	24.71	0.157	0.977	0.081	0.564
8	25.45	0.161	1.000	0.081	0.005
9	25.46	0.159	0.992	0.081	0.206
10	26.39	0.166	1.035	0.080	0.871
11	24.56	0.179	1.111	0.079	2.785
12	25.6	0.174	1.079	0.080	1.992
13	18.91	0.186	1.158	0.079	3.990
300 mL std		0.161		0.057	

Note: Optimized ROI was channels 62 to 171
Note: Total 300 mL gram recovery was 0.996

Table C.5: Distillation 9 Heated on 10 Until after First Vial

Vial	Sample (g)	Activity Concentration (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
1	6.01	0.194	1.106	0.078	2.720
2	25.28	0.172	0.978	0.076	0.582
3	25.1	0.168	0.957	0.076	1.138
4	25.42	0.166	0.945	0.076	1.446
5	25.62	0.159	0.905	0.076	2.500
6	24.76	0.162	0.923	0.076	2.036
7	26.31	0.163	0.927	0.076	1.925
8	27.52	0.174	0.991	0.076	0.240
9	25.83	0.177	1.006	0.076	0.148
10	25.65	0.171	0.976	0.076	0.635
11	25.36	0.178	1.015	0.076	0.408
12	25.08	0.177	1.008	0.075	0.220
13	10.57	0.186	1.062	0.075	1.648
300 mL std		0.176		0.053	

Note: Optimized ROI was channels 62 to 171
Note: Total 300 mL gram recovery was 0.994

Table C.6: Distillation 10 Heated on 8 for Entire Distillation

Vial	Sample (g)	Activity Concentration (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
1	5.71	0.735	0.985	0.040	0.749
2	4.89	0.699	0.937	0.041	3.084
3	21.4	0.662	0.887	0.039	5.828
3+2	26.29	0.674	0.903	0.041	4.720
4	24.59	0.662	0.888	0.039	5.809
5	24.65	0.676	0.906	0.039	4.873
6	25.07	0.665	0.891	0.039	5.665
7	24.99	0.682	0.913	0.039	4.494
8	25.25	0.663	0.889	0.039	5.771
9	25.23	0.677	0.901	0.039	5.115
10	25.13	0.671	0.899	0.039	5.225
11	25.47	0.680	0.911	0.038	4.643
12	25.1	0.702	0.941	0.038	3.049
13	22.64	0.727	0.974	0.038	1.377
14	16.39	0.756	1.014	0.038	0.725
300 mL std		0.746		0.027	

Note: Optimized ROI was channels 60 to 169
Note: Total 300 mL gram recovery was 0.986

Table C.7: Distillation 11 Heated on 8 for Entire Distillation

Vial	Sample (g)	Activity Concentration (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
1	4.9	0.694	0.977	0.040	1.132
2	5.19	0.733	1.031	0.040	1.566
3	20.59	0.643	0.906	0.038	5.004
3+2	25.78	0.673	0.947	0.040	2.635
4	25.27	0.660	0.928	0.038	3.797
5	25.25	0.666	0.937	0.038	3.351
6	25.14	0.671	0.945	0.038	2.912
7	26.29	0.671	0.944	0.038	2.960
8	24.84	0.693	0.975	0.038	1.305
9	24.89	0.693	0.976	0.038	1.272
10	25.07	0.675	0.950	0.037	2.668
11	27.65	0.723	1.017	0.038	0.914
12	25.27	0.716	1.008	0.037	0.405
13	25.31	0.735	1.035	0.037	1.850
14	14.79	0.773	1.089	0.037	4.746
300 mL std		0.710		0.027	

Note: Optimized ROI was channels 60 to 169

Note: Total 300 mL gram recovery was 0.989

Table C.8: Distillation 12 Heated on 8 for Entire Distillation

Vial	Sample (g)	Activity Concentration (Bq/g)	Relative recovery	Relative Uncertainty (k=2)	t-value
1	5.15	0.706	1.004	0.041	0.208
2	5.60	0.711	1.010	0.041	0.511
3	9.94	0.670	0.9536	0.039	2.391
3+2	15.54	0.679	0.966	0.041	1.655
4	10.00	0.655	0.932	0.039	3.479
5	9.86	0.668	0.950	0.039	2.576
6	10.02	0.677	0.963	0.039	1.874
7	9.99	0.669	0.951	0.039	2.516
8	9.85	0.705	1.002	0.039	0.116
9	9.99	0.677	0.963	0.039	1.889
10	10.01	0.680	0.968	0.039	1.654
11	9.98	0.702	0.998	0.039	0.091
12	9.99	0.703	1.000	0.039	0.009
13	10.02	0.728	1.035	0.040	1.804
14	9.98	0.772	1.098	0.039	5.075
300 mL std		0.703		0.028	

Note: Optimized ROI was channels 60 to 169

Note: Total 300 mL gram recovery was 0.993

Appendix D: 20 mL Vial Absorption Data

Table D.1: 20 mL Plastic Vial Relative Recovery for an Activity Concentration of Approximately 0.16 Bq/g

Date Counted	Delay (d)	Activity Concentration* (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
4/3/2012	1	0.223	1.000	0.038	
4/3/2012	1	0.199	0.892	0.055	3.915
4/5/2012	3	0.165	0.741	0.057	9.049
4/5/2012	3	0.185	0.829	0.057	6.059
4/8/2012	6	0.189	0.847	0.056	5.433
4/10/2012	8	0.179	0.804	0.057	6.902
5/10/2012	38	0.184	0.824	0.057	6.194
6/4/2012	63	0.187	0.838	0.057	5.694

* Corrected for decay

Note: ROI was channels 60 to 170

Table D.2: 20 mL Plastic Glass Relative Recovery for an Activity Concentration of Approximately 0.16 Bq/g

Date Counted	Delay (d)	Activity Concentration* (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
4/3/2012	1	0.268	1.000	0.036	
4/3/2012	1	0.278	1.040	0.039	1.777
4/5/2012	3	0.263	0.982	0.039	1.081
4/6/2012	3	0.264	0.988	0.039	0.825
4/8/2012	6	0.266	0.996	0.039	0.487
4/10/2012	8	0.275	1.026	0.039	1.198
5/10/2012	38	0.257	0.964	0.039	2.161
6/4/2012	63	0.260	0.980	0.039	1.680

* Corrected for decay

Note: Optimized ROI was channels 60 to 170

Table D.3: 20 mL Plastic Vial Relative Recovery for an Activity Concentration of Approximately 0.67 Bq/g

Date Counted	Delay (d)	Activity Concentration* (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
4/2/2012	1	0.766	1.000	0.027	
4/3/2012	1	0.919	1.200	0.038	11.472
4/4/2012	3	0.733	0.958	0.039	2.075
4/5/2012	3	1.393	1.819	0.038	42.041
4/7/2012	6	0.676	0.883	0.039	5.761
4/9/2012	8	0.703	0.918	0.039	4.114
5/10/2012	38	0.728	0.950	0.039	2.336
6/4/2012	63	0.689	0.900	0.039	4.762

* Corrected for decay

Note: Optimized ROI was channels 60 to 170

Table D.4: 20 mL Glass Vial Relative Recovery for an Activity Concentration of Approximately 0.67 Bq/g

Date Counted	Delay (d)	Activity Concentration* (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
4/2/2012	1	0.789	1.000	0.027	
4/3/2012	1	0.772	0.979	0.038	1.082
4/4/2012	3	0.782	0.992	0.038	0.435
4/5/2012	3	0.777	0.985	0.038	0.805
4/7/2012	6	0.798	1.011	0.038	0.586
4/9/2012	8	0.791	1.003	0.038	0.145
5/10/2012	38	0.769	0.975	0.039	1.313
6/4/2012	63	0.779	0.987	0.039	0.653

* Corrected for decay

Note: Optimized ROI was channels 60 to 170

Table D.5: 20 mL Plastic Vial Relative Recovery for an Activity Concentration of Approximately 18.6 Bq/g

Date Counted	Delay (d)	Activity Concentration* (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
4/2/2012	1	18.790	1.000	0.022	
4/3/2012	1	18.884	1.005	0.032	0.314
4/4/2012	3	18.889	1.005	0.032	0.332
4/5/2012	3	18.760	0.998	0.032	0.101
4/7/2012	6	18.860	1.004	0.032	0.236
4/9/2012	8	18.921	1.007	0.032	0.440
5/10/2012	38	18.933	1.008	0.032	0.481
6/4/2012	63	18.864	1.004	0.032	0.249

* Corrected for decay

Note: Optimized ROI was channels 60 to 170

Table D.6: 20 mL Glass Vial Relative Recovery for an Activity Concentration of Approximately 18.6 Bq/g

Date Counted	Delay (d)	Activity Concentration* (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
4/3/2012	1	18.969	1.000	0.022	
4/3/2012	1	18.990	1.001	0.037	0.058
4/4/2012	3	18.955	0.999	0.032	0.047
4/5/2012	3	18.868	0.995	0.032	0.336
4/7/2012	6	18.957	0.999	0.032	0.042
4/10/2012	8	18.943	0.999	0.032	0.088
5/10/2012	38	18.975	1.000	0.032	0.019
6/4/2012	63	19.007	1.002	0.032	0.125

* Corrected for decay

Note: Optimized ROI was channels 60 to 170

Appendix E: 1000 mL HDPE Bottle Absorption

Table E.1: 1000 mL HDPE Bottle Absorption Test Data for the Backgrounds

Description	Counted	Activity Concentration (Bq/g)	Relative Uncertainty (k=2)
DI background	3/17/2012	0.187	0.055
DI background	3/17/2012	0.169	0.055
	Average	0.178	0.055

Note: Optimized ROI was channels 60 to 171

Table E.2: 1000 mL HDPE Bottle Absorption Data for Test #1 for an Approximate Activity Concentration of 0.16 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	3/17/2012	0.123	0.142	1.000	0.159	
3/13/2012	3/18/2012	0.123	0.142	1.001	0.158	0.006
3/13/2012	3/18/2012	0.120	0.138	0.973	0.160	0.240
3/14/2012	3/18/2012	0.120	0.139	0.977	0.160	0.201
3/14/2012	3/18/2012	0.120	0.138	0.971	0.160	0.261
3/15/2012	3/18/2012	0.147	0.168	1.187	0.152	1.700
3/17/2012	3/18/2012	0.149	0.171	1.206	0.151	1.880
3/17/2012	5/16/2012	0.106	0.137	0.966	0.159	0.302
3/19/2012	5/16/2012	0.115	0.147	1.039	0.154	0.351
3/22/2012	5/16/2012	0.112	0.143	1.008	0.157	0.070
3/26/2012	5/16/2012	0.119	0.150	1.059	0.154	0.529
4/1/2012	5/17/2012	0.113	0.142	1.000	0.157	0.003

Note: Optimized ROI was channels 60 to 171

* Corrected for decay

Table E.3: 1000 mL HDPE Bottle Absorption Data for Test #2 for an Approximate Activity Concentration of 0.16 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	3/18/2012	0.123	0.142	1.000	0.183	
3/13/2012	3/18/2012	0.117	0.135	0.947	0.187	0.405
3/13/2012	3/19/2012	0.117	0.135	0.951	0.186	0.376
3/14/2012	3/19/2012	0.122	0.141	0.991	0.183	0.073
3/14/2012	3/19/2012	0.124	0.143	1.001	0.183	0.010
3/15/2012	3/19/2012	0.134	0.155	1.086	0.179	0.671
3/17/2012	3/19/2012	0.138	0.158	1.113	0.177	0.890
3/17/2012	5/18/2012	0.113	0.145	1.022	0.179	0.173
3/19/2012	5/18/2012	0.106	0.137	0.959	0.184	0.313
3/22/2012	5/19/2012	0.113	0.144	1.014	0.181	0.112
3/26/2012	5/19/2012	0.109	0.139	0.976	0.184	0.186
4/1/2012	5/19/2012	0.120	0.150	1.056	0.178	0.442

Note: Optimized ROI was channels 60 to 171
* Corrected for decay

Table E.4: 1000 mL HDPE Bottle Absorption Data for Test #3 for an Approximate Activity Concentration of 0.16 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	3/20/2012	0.119	0.137	1.000	0.161	
3/13/2012	3/20/2012	0.116	0.134	0.977	0.162	0.198
3/13/2012	3/20/2012	0.108	0.125	0.910	0.166	0.773
3/14/2012	3/20/2012	0.122	0.141	1.029	0.159	0.259
3/14/2012	3/20/2012	0.113	0.131	0.953	0.163	0.407
3/15/2012	3/21/2012	0.113	0.130	0.947	0.164	0.457
3/17/2012	3/21/2012	0.126	0.144	1.050	0.160	0.443
3/19/2012	3/21/2012	0.136	0.156	1.135	0.155	1.204
3/22/2012	5/26/2012	0.112	0.145	1.056	0.157	0.494
3/26/2012	5/26/2012	0.108	0.140	1.016	0.159	0.146
4/1/2012	5/26/2012	0.108	0.138	1.005	0.160	0.044

Note: Optimized ROI was channels 60 to 171
* Corrected for decay

Table E.5: 1000 mL HDPE Bottle Absorption Data for Test #4 for an Approximate Activity Concentration of 0.37 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	3/22/2012	0.300	0.305	1.000	0.086	
3/13/2012	3/22/2012	0.306	0.311	1.020	0.086	0.323
3/13/2012	3/22/2012	0.302	0.307	1.008	0.086	0.127
3/14/2012	3/22/2012	0.295	0.300	0.983	0.087	0.282
3/14/2012	3/22/2012	0.311	0.316	1.036	0.086	0.585
3/15/2012	3/22/2012	0.292	0.296	0.971	0.087	0.468
3/17/2012	3/22/2012	0.303	0.307	1.005	0.087	0.082
3/19/2012	3/23/2012	0.318	0.320	1.049	0.086	0.806
3/19/2012	5/26/2012	0.303	0.348	1.140	0.082	2.349
3/22/2012	5/26/2012	0.292	0.332	1.090	0.083	1.498
3/26/2012	5/26/2012	0.303	0.343	1.124	0.083	2.082
4/1/2012	5/27/2012	0.299	0.334	1.096	0.083	1.596

Note: Instrument uncertainty calculated by averaging instrument uncertainties of 0.16 Bq/g and 0.67 Bq/g tests.

Note: Optimized ROI was channels 60 to 171

* Corrected for decay

Table E.6: 1000 mL HDPE Bottle Absorption Data for Test #5 for an Approximate Activity Concentration of 0.37 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	3/22/2012	0.299	0.304	1.000	0.086	
3/13/2012	3/22/2012	0.300	0.305	1.003	0.087	0.041
3/13/2012	3/22/2012	0.291	0.296	0.973	0.087	0.434
3/14/2012	3/22/2012	0.306	0.311	1.024	0.086	0.387
3/14/2012	3/22/2012	0.303	0.308	1.012	0.086	0.198
3/15/2012	3/22/2012	0.307	0.311	1.024	0.086	0.391
3/17/2012	3/23/2012	0.304	0.307	1.010	0.087	0.157
3/19/2012	3/23/2012	0.309	0.311	1.023	0.087	0.380
3/22/2012	3/23/2012	0.313	0.313	1.030	0.086	0.493
3/26/2012	5/27/2012	0.297	0.336	1.103	0.083	1.725
4/1/2012	5/27/2012	0.301	0.337	1.107	0.083	1.796

Note: Instrument uncertainty calculated by averaging instrument uncertainties of 0.16 Bq/g and 0.67 Bq/g tests.

Note: Optimized ROI was channels 60 to 171

* Corrected for decay

Table E.7: 1000 mL HDPE Bottle Absorption Data for Test #6 for an Approximate Activity Concentration of 0.37 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	3/26/2012	0.306	0.315	1.000	0.085	
3/13/2012	3/26/2012	0.308	0.316	1.004	0.085	0.069
3/13/2012	3/26/2012	0.300	0.308	0.979	0.085	0.342
3/14/2012	3/27/2012	0.298	0.306	0.972	0.086	0.466
3/14/2012	3/27/2012	0.297	0.304	0.968	0.086	0.533
3/15/2012	3/27/2012	0.294	0.301	0.956	0.086	0.719
3/17/2012	3/27/2012	0.300	0.306	0.972	0.086	0.465
3/19/2012	3/27/2012	0.291	0.296	0.941	0.087	0.972
3/22/2012	3/27/2012	0.295	0.298	0.948	0.086	0.863
3/26/2012	3/27/2012	0.312	0.313	0.995	0.086	0.087
3/26/2012	5/27/2012	0.297	0.336	1.068	0.082	1.157
4/1/2012	5/27/2012	0.304	0.340	1.080	0.082	1.348

Note: Instrument uncertainty calculated by averaging instrument uncertainties of 0.16 Bq/g and 0.67 Bq/g tests.

Note: Optimized ROI was channels 60 to 171

* Corrected for decay

Table E.8: 1000 mL HDPE Bottle Absorption Data for Test #7 for an Approximate Activity Concentration of 0.67 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	3/28/2012	0.493	0.513	1.000	0.063	
3/13/2012	3/28/2012	0.501	0.521	1.015	0.063	0.345
3/13/2012	3/28/2012	0.510	0.531	1.033	0.062	0.756
3/14/2012	3/28/2012	0.508	0.529	1.030	0.062	0.685
3/14/2012	3/28/2012	0.494	0.514	1.001	0.063	0.022
3/15/2012	3/29/2012	0.499	0.519	1.012	0.063	0.262
3/17/2012	3/29/2012	0.504	0.524	1.021	0.062	0.472
3/19/2012	3/29/2012	0.500	0.520	1.012	0.063	0.272
3/22/2012	3/29/2012	0.497	0.517	1.007	0.063	0.168
3/26/2012	3/29/2012	0.506	0.527	1.026	0.062	0.586
4/1/2012	5/27/2012	0.494	0.514	1.001	0.064	0.013

Note: Optimized ROI was channels 60 to 171

* Corrected for decay

Table E.9: 1000 mL HDPE Bottle Absorption Data for Test #8 for an Approximate Activity Concentration of 0.67 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	5/12/2012	0.572	0.645	1.000	0.046	
3/13/2012	5/12/2012	0.571	0.644	0.998	0.046	0.050
3/13/2012	5/13/2012	0.569	0.642	0.995	0.046	0.140
3/14/2012	5/13/2012	0.583	0.657	1.018	0.046	0.556
3/14/2012	5/13/2012	0.582	0.656	1.017	0.046	0.521
3/15/2012	5/13/2012	0.569	0.640	0.992	0.046	0.231
3/17/2012	5/13/2012	0.577	0.646	1.002	0.046	0.056
3/19/2012	5/13/2012	0.600	0.669	1.038	0.046	1.171
3/22/2012	5/13/2012	0.574	0.637	0.987	0.046	0.396
3/26/2012	5/13/2012	0.611	0.672	1.042	0.046	1.302
4/1/2012	5/13/2012	0.582	0.633	0.982	0.047	0.540

Note: Optimized ROI was channels 60 to 171
* Corrected for decay

Table E.10: 1000 mL HDPE Bottle Absorption Data for Test #9 for an Approximate Activity Concentration of 0.67 Bq/g

Date Sampled	Date Counted	Activity Concentration* (Bq/g)	Stability Corrected (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
3/13/2012	5/14/2012	0.593	0.671	1.000	0.044	
3/13/2012	5/14/2012	0.605	0.685	1.020	0.044	0.651
3/13/2012	5/14/2012	0.587	0.665	0.991	0.045	0.298
3/14/2012	5/14/2012	0.608	0.687	1.024	0.044	0.762
3/14/2012	5/15/2012	0.601	0.679	1.012	0.044	0.388
3/15/2012	5/15/2012	0.601	0.679	1.011	0.045	0.356
3/17/2012	5/15/2012	0.591	0.665	0.991	0.045	0.288
3/19/2012	5/15/2012	0.600	0.672	1.002	0.045	0.067
3/22/2012	5/15/2012	0.586	0.652	0.972	0.045	0.883
3/26/2012	5/15/2012	0.591	0.653	0.973	0.045	0.861
4/1/2012	5/15/2012	0.574	0.626	0.933	0.046	2.089

Note: Optimized ROI was channels 60 to 171
* Corrected for decay

Appendix F: 1000 mL HDPE bottle desorption

Table F.1: 1000 mL HDPE Bottles Desorption Initial

Description	Activity Concentration (Bq/g)	Relative Uncertainty (k=2)
DI background	0.074	0.030
DI background	0.070	0.030
DI background	0.074	0.030

Note: Optimized ROI was channels 60 to 170

Table F.2: 1000 mL HDPE Bottles Desorption Week 1

First Week	Activity Concentration* (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
Bottle 1	0.112	1.533	0.043	10.193
Bottle 2	0.094	1.289	0.043	5.507
Bottle 3	0.101	1.389	0.043	7.406
Bottle 4	0.097	1.329	0.043	6.261
Bottle 5	0.085	1.158	0.043	3.011
Bottle 6	0.083	1.143	0.043	2.726
Bottle 7	0.081	1.112	0.043	2.132
Bottle 8	0.075	1.031	0.043	0.589
Bottle 9	0.080	1.102	0.043	1.943

Note: Optimized ROI was channels 60 to 170

* Corrected for decay

Table F.3: 1000 mL HDPE Bottles Desorption Week 2

Second Week	Activity Concentration* (Bq/g)	Relative Recovery	Relative Uncertainty (k=2)	t-value
Bottle 1	0.067	0.915	0.039	1.790
Bottle 2	0.074	1.010	0.039	0.201
Bottle 3	0.069	0.939	0.039	1.278
Bottle 4	0.068	0.926	0.039	1.547
Bottle 5	0.066	0.905	0.039	1.990
Bottle 6	0.071	0.974	0.039	0.545
Bottle 7	0.075	1.021	0.039	0.448
Bottle 8	0.074	1.020	0.039	0.414
Bottle 9	0.070	0.962	0.039	0.798

Note: Optimized ROI was channels 60 to 170

* Corrected for decay