THESIS

INVESTIGATION OF TRITIUM ATOM EXCHANGE IN PLASTIC LIQUID SCINTILLATION VIALS

Submitted by

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ABSTRACT

INVESTIGATION OF TRITIUM ATOM EXCHANGE IN PLASTIC LIQUID SCINTILLATION VIALS

Tritium is a naturally occurring radionuclide; it is an analyte of interest in many air, soil, and water samples. It has been shown that long term storage and study of tritium samples results in a reduction in tritium activity not attributed to the natural radioactive decay. Several explanations have been offered through past literatures including diffusion, LSC cocktail degradation, and change in quenching effects. Another possible explanation for the decrease in activity is that tritium may have been organically bound to the plastic possibly due to exchangeable hydrogen atoms along the plastic carbon chain. The hypothesis that tritium can be incorporated into the plastic, interchanging the ¹H atoms in the plastic with ³H atoms, was experimentally tested. The experiment consisted of adding deionized water into a previously used plastic vial which had contained tritium to determine if the deionized water had now become tritiated. The results showed that the longer the tritiated water is stored in the vials, the greater the loss of tritium activity in plastic vials is compared to glass vials. An increase in the time that the tritiated water is stored also increases the activity of the tritium found in the deionized water in plastic vials but not in the glass vials. The combination of these two observations supports the hypothesis that tritium exchange may have occurred between the tritiated water and the hydrogen within the plastic vials.

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CHAPTER 1 INTRODUCTION

Tritium, an isotope of hydrogen, is a naturally occurring cosmogenic radionuclide. One of the most common ways that tritium is formed is in the atmosphere by the bombardment of fast neutrons from cosmic rays onto ¹⁴N (Nir, Kruger, Lingenfelter & Flamm, 1966). The reaction is shown in equation 1.1:

$$^{14}N + n^{o} \rightarrow ^{15}N \rightarrow ^{12}C + ^{3}H.$$
 (1.1)

The production of tritium in the atmosphere yields an activity of approximately 4 megacuries, or 148,000 terabecquerel, per year with a natural equilibrium level of 70 megacuries (NCRP 62, 1979). Curie (Ci) and becquerel (Bq) are both units of radioactivity; Bq is the unit in the International System of Units (SI) for radioactivity whereas Curie is the unit in the non-SI units used in the United States; they represent the number of decays or disintegrations per second. In addition to the natural tritium production in the atmosphere, the surface nuclear weapons testing during World War II and the Cold War increased the tritium inventory in the atmosphere to a maximum of 3100 megacuries (NCRP 62, 1979). Since tritium still has one proton and one electron, for the most part, it behaves chemically like normal hydrogen, ¹H. In any compounds containing hydrogen, ¹H, a ¹H atom can theoretically be replaced with tritium; for example, one common tritium compound is tritiated water, commonly designated as HTO (NCRP 62, 1979 & ICRP 119, 2012). HTO is water with one of the ¹H replaced by ³H. In the atmosphere, tritium can exist in water vapor and after several reactions it can subsequently be precipitated onto the earth's surface, which contributes to the natural background radiation found in our water system (NCRP 62). The abundance of tritium in nature has prompted researchers to study tritium in the environment since the 1950's, which highlights the need for accurate counting.

Tritium is composed of one proton and two neutrons. Compared to stable hydrogen and deuterium, 1 H and 2 H, tritium has excess neutrons. As a result, tritium will undergo beta-minus decay, which essentially converts one of its neutrons to a proton as depicted in Figure 1. Along with the emission of a beta particle, all beta-minus decay will also emit an antineutrino. The kinetic energy of the beta-minus decay is then split between the beta particle and the antineutrino, yielding the polyenergenetic nature of the beta particles. For tritium, the beta particles have a mean energy of 5.68 keV and a maximum energy of 18.591 keV, with a half-life of 12.32 years (Baum, 2009). The beta particles emitted from tritium do not travel very far at these energies; for example, the range in air is 4.5 to 6 mm, the range in water is 5 μ m, and in tissue it is approximately 7 μ m (U.S. Department of Energy, 1999). The short ranges of these particles limit the useable detection instruments as most instruments will block these beta particles before reaching the detection medium. As a result, the most common and reliable way to detect tritium is through Liquid Scintillation Counting (LSC) (Health Physics Society, 2011).

Tritium Beta-minus Decay

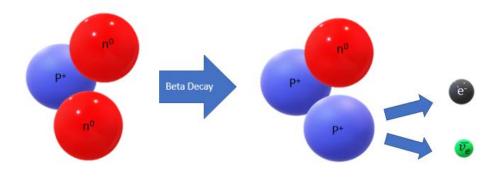


Figure 1. Schematic of Tritium Beta Minus Decay

Liquid Scintillation Counting follows the same concept as any scintillation counters. As ionizing radiation interacts with the scintillation material, it deposits its energy and the scintillator reaches an excited state and de-excites by the emission of light; this light can then be counted and measured to quantify ionizing radiation. The main advantage of liquid scintillation counting is the utilization of liquid scintillation cocktails that can be mixed directly with the sample to reduce the distance traveled by the ionizing radiation. This is particularly useful for low energy beta emitting radionuclides such as tritium and carbon-14. The liquid scintillator mixture can contain both organic and/or inorganic material and there are multiple commercial products that are used for different counting applications (Kessler, 2015). These liquid scintillator mixtures are often referred to as LSC cocktail. One advantage of scintillation counting is that the intensity of the light emitted is proportional to the energy of the ionizing radiation (Kessler, 2015). This trait allows the scintillation detector to discriminate different energies and act as a spectrometer. A common tritium energy spectrum is shown in Figure 2. The shape of the peak is common among beta decays, the peak is a classic representation of the polyenergetic characteristics of beta particles. The peak also appears on the left side of the spectrum, indicating the low energy characteristics of the tritium beta particles.

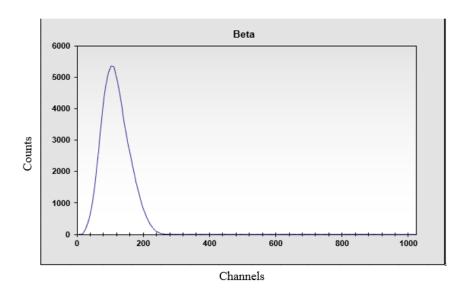


Figure 2. Tritium standard spectrum obtained using Hidex 300SL

Liquid scintillation counting of tritium has been utilized for several decades, as a result, past publications have identified several factors that may affect accurate background determination in LSC. One such factor is the effect of phosphorescence on the LSC cocktail. Phosphorescence occurs when the LSC cocktail is exposed to sunlight which excites the scintillation solution. In 1969, the effects of phosphorescence were observed to persist as long as 70 hours before dissipating (Moghissi, Kelley, Regnier & Carter, 1969). The effect of phosphorescence on the LSC cocktail can be reduced by changing the composition of LSC cocktail and avoid direct sunlight irradiations (Moghissi, Kelley, Regnier & Carter, 1969). The second factor that affects liquid scintillation counting is the chemiluminescence property of the LSC cocktail. Chemiluminescence is the emission of light during a chemical reaction (Welsh, 2011). Past research has demonstrated that the mixing of the LSC cocktail and the polyethylene vial yielded increased sample counts, which were attributed to the effects to chemiluminescence (Moghissi, Kelley, Regnier & Carter, 1969). Lastly, the effects of static electricity on the LSC background count have been observed by past researchers; this effect was more prominent when the sample had been automatically moved within the chamber multiple times (Moghissi, Kelley, Regnier &

Carter, 1969). The use of anti-static solution on the scintillation vials has proved to reduce the effects of static electricity on LSC with no observable effects on the efficiency of the counting instrument (Moghissi, Kelley, Regnier & Carter, 1969). These effects have since been reduced by improving the methods and materials used for LSC measurements for an accurate background measurement.

In the late 1960's, a concept known as the "figure of merit" or FOM was introduced. This value is calculated by using the equation

$$FOM = \frac{E^2}{B}, \tag{1.2}$$

where E is the efficiency in % and B is the background in cpm; the FOM is a value used to determine the signal to background ratio or another term, "background dominant determination" (Moghissi, Kelley, Regnier & Carter, 1969). This value is useful when comparing different counters with low or high background counts (Moghissi, Kelley, Regnier & Carter, 1969). The FOM has since then been adopted by LSC manufacturers to determine the optimized counting region for a radionuclide for the instrument, as FOM can also represent counting sensitivity (Passo & Cook, 2002). By adjusting the counting regions or channels and calculating the FOM, the highest FOM signifies the optimized window for a specific radionuclide against background.

One important concept in LSC measurements is the effect of quenching; a phenomenon that can significantly reduce the counting efficiency (Jakonić, Nikolov, Todorović & Vesković, 2014). There are four different types of quenching: "absorption or physical quenching, chemical quenching, photon or color quenching and solvent dilution quenching (Jakonić, Nikolov, Todorović & Vesković, 2014)." Essentially, absorption or physical quenching and photon or color quenching effects prevent the emitted light due the radiation interactions from reaching the

photomultiplier tubes at its full intensity, while chemical quenching and solvent dilution quenching may prevent radiation energy transfer from a solvent molecule to the scintillating solute molecule, effectively preventing light emission. These quenching effects may reduce the counting efficiency or potentially shift the energy peaks (Kessler, 2015). To account for the quenching effects, standard procedures utilize quenching curves to perform quenching corrections. Quenching curves are generated by plotting a quench indicating parameter (QIP) against its associated efficiency and then matching the quench of the sample to adjust or correct the efficiency of the detector (Kessler, 2015). The four main types of QIP are: sample spectrum QIPs, external standard spectrum QIPs, internal standardization, and efficiency tracing DPM (Kessler, 2015). An example of a quench curve can be found in Figure 3. Properly accounting for the quenching effects of a sample is an important process for liquid scintillation counting, it must be carefully researched and implemented to obtain an accurate measurement.

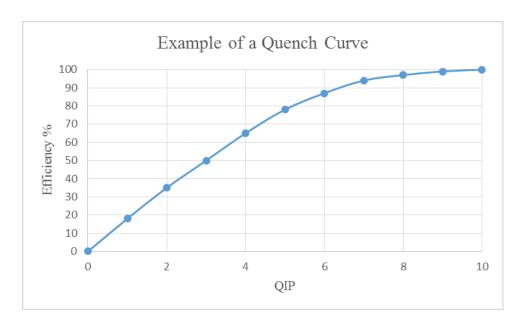


Figure 3. Example of a Quench Curve. The QIP could vary depending on the type of quenching effect the sample is experiencing. The efficiency will also vary depending on the effects from the QIP. Once the curve has been plotted, the efficiency of the instrument can be found be determining the magnitude of the QIP in the sample.

Scintillation technology has improved over the years. One such improvement is the utilization of multiple photomultiplier detectors. Since the mid 1980's, a double photomultiplier detector system has been used to eliminate thermal background and afterpulses (Pochwalski, Broda & Radoszewski, 1986). Some modernized systems now utilize triple-photomultiplier liquid scintillation detectors, depicted in Figure 4, with two different coincident outputs, which is often referred to as the TDCR or triple-to-double coincidence ratio technology (Pochwalski, Broda & Radoszewski, 1986). TDCR instruments work by comparing the triple coincidence to double coincidence of the photomultiplier tubes (Pochwalski, Broda & Radoszewski, 1986). "The under-lying theory is based on a statistical distribution law of the number of photons emitted by the scintillation process, (Priya, Gopalakrishnan & Goswami, 2014)." In recent studies, the TDCR for an instrument has been shown to approximate the overall counting efficiency for the Hidex 300SL when analyzing pure beta-emitting radionuclides (Priya, Gopalakrishnan & Goswami, 2014). This estimated efficiency calculated from TDCR is independent of the quenching effect, hence eliminating the need for quenching curves. TDCR technology has been developed to eliminate the cost of quenching standards, to minimize the radioactive waste generated from making quenching standards, and to reduce the time required to generate quenching curves.

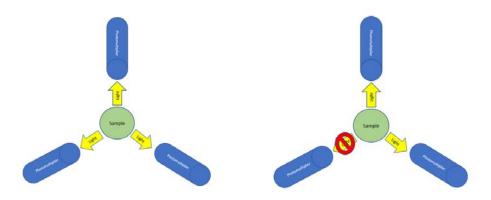


Figure 4. Example of a Triple Photomultiplier LSC Chamber. The left figure depicts an unobstructed light emission where the figure to the right have obstruction of the light, due to quenching effects.

As with all samples gathered from a population, statistical analysis is required to accurately characterize the true data. Statistical analysis is even more critical in the field of Health Physics since radiation decay is a stochastic or random process. A sample measurement, in addition to the radioactive decay that may affect its counts, will be subject to fluctuation. This fluctuation and the random nature of radiation fits the normal distribution model, which means that the "true" count is really the "true average" count (Johnson, 2017). This random behavior also applies to background counts. An instrument remaining in the same location, counting background for the same amount of time, can have different measurement results where the combination of such results will follow a normal distribution. The normal distribution is a common statistical model that fits many natural processes; it essentially describes the variations that occur naturally (Wackerly, Mendenhall & Scheaffer, 2008). Another common distribution used in measuring radioactivity is the Poisson distribution. The Poisson distribution is a good model for events with low chance of occurrence. It is useful for describing natural decay process because the probability of a single atom decaying is small (Wackerly, Mendenhall & Scheaffer, 2008; Johnson, 2017). As the number of decays or counts increases, the Poisson distribution behaves more like the normal distribution (Johnson, 2017). The Poisson distribution is

commonly used in operational health physics, especially when only one measurement is utilized, because the standard deviation of a Poisson distributed mean is the square root of the mean (Johnson, 2017). These distributions describe the natural variation and the randomness of the radioactive decays. Selecting and performing the appropriate statistical analysis is important in distinguishing between the subtle increase in counts and the spread of these distributions.

One powerful statistical tool for health physicists is the t-test. The t-test is appropriate especially for analysis of small samples (Wackerly, Mendenhall & Scheaffer, 2008). The t-test is a hypothesis test that is used to compare two sample means. Hypothesis testing has four elements: 1. The null hypothesis, 2. The alternative hypothesis, 3. Test statistic and 4. The rejection region (Wackerly, Mendenhall & Scheaffer, 2008). The null hypothesis is the hypothesis that is to be tested; in health physics application, this hypothesis is usually the following: the mean of the sample is equal to the mean of the background. Next, the alternative hypothesis is the hypothesis that a researcher is trying to support; an example of an alternative hypothesis is that the sample mean is greater than the mean of the background (Wackerly, Mendenhall & Scheaffer, 2008). The test statistic is a calculated value that can be used to compare against the rejection region to determine whether to accept or reject the null hypothesis (Wackerly, Mendenhall & Scheaffer, 2008). The rejection region must be carefully chosen, it is based on either the type I error, type II error or both (Wackerly, Mendenhall & Scheaffer, 2008). Type I error, denoted by α , is made when the null hypothesis is rejected when it is true whereas the type II error, denoted by β , is made when the null hypothesis is accepted when the alternative hypothesis is true (Wackerly, Mendenhall & Scheaffer, 2008). These two errors are important concepts in health physics, as they are referred to as false alarm and missed detection (Johnson,

2017). In operational health physics, these errors are commonly accepted when set at 5%. To perform the t-test, the following formula is used to calculate the test statistic:

$$t = \frac{|M_1 - M_2|}{\sigma_{\text{diff}}} \tag{1.3}$$

where

$$\sigma_{\text{diff}} = \sqrt{\sigma_{\text{M}_1}^2 + \sigma_{\text{M}_2}^2} \tag{1.4}$$

(Johnson, 2017). Once the t-value is calculated, it can then be compared to the relevant rejection region. The t-value on its own can also be indicative of how big the difference between the two means is while taking account of the standard deviation of the mean. The bigger the magnitude of the t-value means that the difference in means is larger while accounting for statistical fluctuation. The utilization of a t-test is not only important but necessary in determining the presence of a small amount of radioactive material in a sample.

Tritium is a naturally occurring radioactive material from the atmosphere; it travels to the Earth's surface via rain and snow and follows where water would go. Logically, it is not unusual to find tritium in natural water. The need to distinguish between naturally occurring tritium and man-made tritium contamination in an environmental water sample can be of interest for public safety and regulators. LSC counting of tritium in water is common practice such as described by Environmental Protection Agency Method 906.0. This method recommends both glass and polyethylene vials as containers for the LSC vial depending on the LSC cocktail used (U.S. EPA, 1980). One advantage of polyethylene vials over regular glass vials is that polyethylene vials can yield a lower background as regular glass vials can contain naturally occurring potassium-40 atoms (40 K). 40 K undergoes beta-minus decay 89.28% of the time; it has a mean beta energy of

560.2 keV with a maximum beta energy of 1310.89 keV. Without using the optimized window, the fluctuating amount of ⁴⁰K in a vial can influence counting results. Therefore, low-potassium glass vials or polyethylene vials are recommended for tritium counting. However, past research has shown that long term storage of tritium in plastic vials can result in counting rate losses in addition to the natural radioactive decay (Nedjadi, Duc, Bochud & Bailat, 2016). Several factors have been identified as possible reasons for the counting rate losses: the stability of the scintillation cocktail, diffusion of the scintillation cocktail and sample through the polyethylene vials and long-term quenching changes within a sample (Nedjadi, Duc, Bochud & Bailat, 2016; Verrezen, Loots & Hurtgen, 2008; Feng, He, Wang & Chen, 2015). One additional hypothesis was proposed as a cause for the counting rate loss. Recall that tritium can exchange places with ¹H, theoretically, tritium can replace the ¹H found on the polyethylene carbon chain. If enough tritium bonded within the porous section of the plastic, when it decays, it may be subject to selfabsorption before reaching the LSC cocktail, resulting in a loss of count. In a simple experiment performed by a previous CSU graduate student, tritiated water was allowed to soak in a plastic gallon container. The container was then dried, and deionized water was added to it. The deionized water was sampled and counted in the LSC, yielding an increased count in the optimized region for tritium. The experimental result showed that tritium was introduced to the deionized water within the dried plastic container; the unknown source of the tritium could be the result of ¹H and ³H atom exchange along the polyethylene carbon chain. The main objective of the following experiment is to test the hypothesis that tritium atom exchange can occur within plastic LSC vials and as a result, reduce radioactivity measurements.

CHAPTER 2 MATERIALS AND METHODS

The following experiment is designed to determine if tritium could interchange with the hydrogen atoms that are present in the organic polymers found in the typical plastic LSC vials. It mirrors the simple experiment mentioned at the end of Chapter 1, but with additional controls to isolate variables. The main premise of the experiment is to soak a clean LSC vial with tritiated water for a predetermined amount of time, clean the vial via drying, and soak the dried vial with deionized water to see if there was any tritium being reabsorbed by the water. Both plastic and glass vials were used for comparison purpose. DWK Life Sciences (Kimble) Polyethylene 20 ml LSC vials were chosen for the plastic vials, and Wheaton Disposable Scintillation glass 20 ml LSC vials with foil lined caps were selected for the glass vials.

The experiment is broken up into two separate parts, with the major distinction being the tritiated water soaking time. In this experiment, soaking time is defined as the amount of time that a LSC vial is in contact with NIST traceable tritiated water. The first part allowed soaking times between one to seven days, while the second part utilized soaking times between 7 days through 84 days. The soaking times are displayed in Table 1.

Table 1. Tritiated Water Soaking Times

Tritiated Water Soaking Times										
Part I 1 day 2 days 3 days 4 days 5 days 6 days 7 days										
Part II	7 days	10 days	14 days	21 days	35 days	56 days	84 days			

Five plastic vials and five glass vials were used for each soaking time duration. For each set of five, four were soaked in 8 mL of NIST traceable tritiated water with an activity of 2400 Bq/mL and the fifth vial was soaked in 8 mL of deionized water as control. The naming

convention for the vials is listed in Table 2 and Table 3. The vials were weighed before and after adding their respective liquid to determine the volume of liquid added.

Table 2. Sample Labeling Naming Convention for Part I Experiment

Part 1	I												
Da	ay 1	Da	ıy 2	Da	y 3	Da	y 4	Da	y 5	Da	y 6	Da	y 7
1P1	1G1	2P1	2G1	3P1	3G1	4P1	4G1	5P1	5G1	6P1	6G1	7P1	7G1
1P2	1G2	2P2	2G2	3P2	3G2	4P2	4G2	5P2	5G2	6P2	6G2	7P2	7G2
1P3	1G3	2P3	2G3	3P3	3G3	4P3	4G3	5P3	5G3	6P3	6G3	7P3	7G3
1P4	1G4	2P4	2G4	3P4	3G4	4P4	4G4	5P4	5G4	6P4	6G4	7P4	7G4
1PB	1GB	2PB	2GB	3PB	3GB	4PB	4GB	5PB	5GB	6PB	6GB	7PB	7GB

Table 3. Sample Labeling Naming Convention for Part II Experiment

Part I	I												
Da	ay 7	Da	y 10	Day	/ 14	Day	y 21	Day	y 35	Day	y 56	Day	y 84
7-2P1	7-2G1	10P1	10G1	14P1	14G1	21P1	21G1	35P1	35G1	56P1	56G1	84P1	84G1
7-2P2	7-2G2	10P2	10G2	14P2	14G2	21P2	21G2	35P2	35G2	56P2	56G2	84P2	84G2
7-2P3	7-2G3	10P3	10G3	14P3	14G3	21P3	21G3	35P3	35G3	56P3	56G3	84P3	84G3
7-2P4	7-2G4	10P4	10G4	14P4	14G4	21P4	21G4	35P4	35G4	56P4	56G4	84P4	84G4
7-2PB	7-2GB	10PB	10GB	14PB	14GB	21PB	21GB	35PB	35GB	56PB	56GB	84PB	84GB

After fulfilling the predetermined soaking time, the contents of the vials were transferred to a set of clean plastic vials. The remaining drops were removed and discarded via disposable pipettes. The transferred contents were labeled as "waste" with the vials labeled following the original samples conventions with an additional W added to it. The waste samples were kept for further analysis; however, this process was not introduced and correctly implemented until Part I, day 4 of the experiment and Part II, day 14 of the experiment, so some of the waste data were lost as missed opportunities or failed data gathering. The now empty vials that had their previous contents removed were then transferred to an Across International vacuum drying oven. The vacuum chamber was purged with nitrogen gas to remove any atmospheric water vapor and the vials were then dried for two days at a setting of 80 degrees Celsius and a vacuum pressure of 630 mmHg. After drying, the vials were weighed prior to adding deionized water and weighed once again after the addition of water for volume determination. The newly added deionized

water underwent a reabsorption time of 7 days, where the objective is to allow tritium that may have been bound to the LSC vials to exchange back with the hydrogen in the deionized water.

For Part I of the experiment, 12 ml of the Ultima Gold LLT (Low Level Tritium) Liquid Scintillation Cocktail was added immediately to each vial after adding the deionized water for counting. The samples were counted again after 7 days. The two sets of measurement methodology attempted to determine if atom exchange would occur in the early or later part of the 7-day reabsorption time. The first set of the measurements was coined as initial measurement or initial count. Due to the time constraints between samples, a five-minute count of the sample was used for the initial measurement. The initial measurement was performed immediately after adding the deionized water and the LSC cocktail to determine if the atom exchange is a fast reaction. After 7 days, the second set of measurements was performed where the samples were counted twice for one hour each. They were counted consecutively in the following order: Plastic Vial 1, Plastic Vial 2, Plastic Vial 3, Plastic Vial 4, Plastic Vial Background, Glass Vial 1, Glass Vial 2, Glass Vial 3, Glass Vial 4, Glass Vial Background and repeated once again in the same order. All samples were counted using the Hidex 300SL Liquid Scintillation Counter with at least 15 minutes of dark adapt process. These sample count data are considered raw data until they have gone through statistical analysis.

For Part II of the experiment, after adding the deionized water post the drying process, the samples were stored for the duration of the reabsorption time prior to adding the LSC cocktail to allow atom exchange to only occur between the deionized water and the vials without the presence of the cocktail. After reaching their respective soaking time, 12 mL of LSC cocktail was added to each sample and transported for measurement. A graphical summary of the experiment can be found in Figure 5.

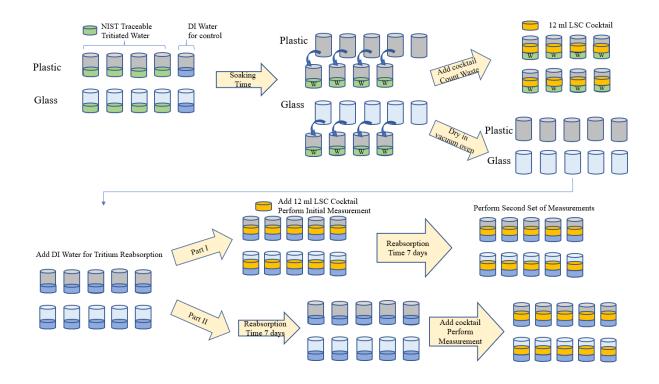


Figure 5. A graphical summary of the experiment

All samples in Part II of the experiment were counted twice for one hour each. They were counted consecutively in the following order: Plastic Vial 1, Plastic Vial 2, Plastic Vial 3, Plastic Vial 4, Plastic Vial Background, Glass Vial 1, Glass Vial 2, Glass Vial 3, Glass Vial 4, Glass Vial Background and repeated once again in the same order. All samples were counted using the Hidex 300SL Liquid Scintillation Counter with at least 15 minutes of dark adapt process. These sample count data are considered raw data until they have gone through statistical analysis. Counting results can be found in the Results section. Results are reported within the optimized window. The optimized window differs between Part I and Part II of the experiment as the counting chamber had to be exchanged due to mechanical issues.

Due to inconsistencies in the data observed in Part II of the experiment, the counting method was modified to five consecutive 30-minute counts per vial. Detailed reasoning is provided in the Results section.

The waste samples gathered prior to the drying process were all immediately measured for their radioactivity via the Hidex 300SL Liquid Scintillation Counter with at least 15 minutes of dark adapt process. All waste samples were counted twice during the measurement at 300 seconds each. As previously mentioned, the waste data gathering procedures were not introduced and correctly implemented until Part I, day 4 of the experiment and Part II, day 14 of the experiment so some of the data were incorrectly gathered or lost as a missed opportunity.

The final control experiment utilized the remaining NIST traceable tritiated water standard. This experiment attempted to characterize the activity of the tritiated water in plastic vials without undergoing the drying process for comparison purpose. The remaining NIST traceable tritiated water yielded 6 vials of 8 ml standard labeled S1-S6. These vials followed a soaking time found in Table 4 prior to adding the LSC cocktail for measurements. These samples were then counted twice for 300 seconds each using the Hidex 300SL Liquid Scintillation Counter with at least 15 minutes of dark adapt process. This aforementioned experiment will be referred to as the "Tritium degradation in plastic vial experiment".

Table 4. Soaking Time for Tritium Degradation in Plastic Vial Experiment

Soaking Time for Tritium Degradation in Plastic Vial Experiment										
Vial	Vial S1 = 10 S2 = 14 S3 = 21 S4 = 35 S5 = 56 S6 = 84									
	days	days	days	days	days	days				

CHAPTER 3 RESULTS

The raw data counts were obtained using the Hidex 300SL Liquid Scintillation Counter along with an Excel macro for visual representation of the data supplied by the manufacturer found in Appendix A. All data reported in this section was measured within the optimized window. The raw counting data for Part I and Part II of the experiment can be found in Table 5 through Table 10.

Part I – Measurement Data

Initial measurement – Immediately after adding fresh deionized water and LSC cocktail

	Plasti	c Vial Gro	oss Counts	s in FOM	75-125		
T01	D.1	TD 4	D2	D 4	D.=	D.(Г

	Plastic Vial Gross Counts in FOM 75-125										
Plastic	D1	D2	D3	D4	D5	D6	D7				
Vial 1	33	27	21	17	30	32	30				
Vial 2	30	22	20	30	35	18	22				
Vial 3	21	28	20	24	30	21	26				
Vial 4	26	33	24	26	22	27	22				
Bkg	23	22	21	20	30	20	26				

Table 5. Part I Experiment Raw Data Initial Count for Plastic Vials

Table 6. Part I Experiment Raw Data Initial Count for Glass Vials

G	Glass Vial Gross Counts in Optimized Window 25-170										
Glass	D1	D2	D3	D4	D5	D6	D 7				
Vial 1	102	126	112	116	101	105	93				
Vial 2	100	131	109	102	127	90	108				
Vial 3	102	107	97	96	97	109	97				
Vial 4	107	101	106	109	106	105	101				
Bkg	100	98	97	103	100	117	117				

7 Days after adding the deionized water and the LSC cocktail

Table 7. Part I Experiment Raw Data Count for Plastic Vials After 7 Days

Plastic Gross Counts in Optimized Window 75-125									
Plastic	Plastic D1 D2 D3 D4 D5 D6 D7								
Vial 1	312	306	345	311	370	188	331		

Vial 2	288	301	320	304	325	195	307
Vial 3	320	309	331	327	268	172	289
Vial 4	310	316	307	299	255	166	301
Bkg	273	299	289	260	304	154	295
Vial 1	296	306	290	275	294	177	310
Vial 2	313	296	319	297	291	201	315
Vial 3	284	288	310	296	291	143	248
Vial 4	255	308	305	317	306	156	285
Bkg	282	271	306	291	279	187	298

^{*}Due to human error, D6 data was set counted for 2017 seconds instead of 3600 seconds resulting in a smaller raw count

Table 8. Part I Experiment Raw Data Count for Glass Vials After 7 Days

	Glass Gross Counts in Optimized Window 25-170							
Glass	D1	D2	D3	D4	D5	D6	D7	
Vial 1	1276	1315	1341	1259	1288	679	1343	
Vial 2	1291	1299	1296	1258	1306	747	1282	
Vial 3	1287	1220	1229	1372	1248	691	1303	
Vial 4	1267	1239	1261	1290	1290	750	1187	
Bkg	1253	1276	1230	1194	1263	778	1266	
Vial 1	1206	1318	1217	1278	1266	705	1303	
Vial 2	1309	1259	1251	1303	1219	723	1197	
Vial 3	1288	1178	1179	1213	1242	712	1242	
Vial 4	1231	1273	1286	1301	1243	704	1302	
Bkg	1286	1317	1368	1260	1284	840	1226	

^{*}Due to human error, D6 data was set counted for 2017 seconds instead of 3600 seconds resulting in a smaller raw count

Part II - Measurement Data

7 Days after adding deionized water

Table 9. Part II Experiment Raw Data Count for Plastic Vials After 7 Days

	Plastic Gross Counts in Optimized Window 21-185							
Plastic	D7	D10	D14	D21	D35	D56	D84	
Vial 1	2204	1096	1746	1278	1468	1563	1885	
Vial 2	3752	1152	3103	6061	7937	1083	1130	
Vial 3	2493	1071	1130	1821	1383	1969	5784	
Vial 4	3613	1170	2780	1207	1132	2140	2093	
Bkg	1665	1099	2424	964	974	892	903	
Vial 1	1788	1095	1158	989	929	939	1067	
Vial 2	1834	1076	1202	1291	1024	936	1127	
Vial 3	1884	1272	2505	1309	1215	1414	1605	

Vial 4	1705	1239	1202	1707	1041	971	1029
Bkg	981	1221	1020	965	5227	1081	1814

Table 10. Part II Experiment Raw Data Count for Glass Vials After 7 Days

	Glass Gross Counts in Optimized Window 18-162							
Glass	D7	D10	D14	D21	D35	D56	D84	
Vial 1	1361	1353	1471	1337	1374	1341	1244	
Vial 2	1457	1415	1428	1269	1349	1305	1282	
Vial 3	1327	1375	1439	2470	1285	1363	1262	
Vial 4	1358	1386	1418	4529	1425	1334	1156	
Bkg	1341	1314	1324	1261	1259	1311	1166	
Vial 1	1444	1381	1483	1306	1288	1308	1202	
Vial 2	1529	1417	1436	1321	1387	1268	1195	
Vial 3	1435	1375	1445	2336	1440	1330	1234	
Vial 4	1377	1362	1383	4587	1483	1272	1195	
Bkg	1386	1370	1422	1246	1216	1281	1183	

Upon examination of the gross count data, the counts of the plastic vials, and only the plastic vials, displayed a strange inconsistency. For example, for vial 35P2 (35 days of tritiated water soaking, sample vial 2) the two counts of the same vial yielded 7937 and 1024 counts. For a 1-hour count, the difference is concerning. By examining the spectra of the plastic vials shown in Figure 6, the peak, or the increased counts, between channels 0-200 appeared sporadically within a same vial. This inconsistency is not a normal characteristic of tritium's beta radiation. Since these peaks are within the tritium optimized window for plastic, further investigation was warranted to determine the source of these sporadic peaks.

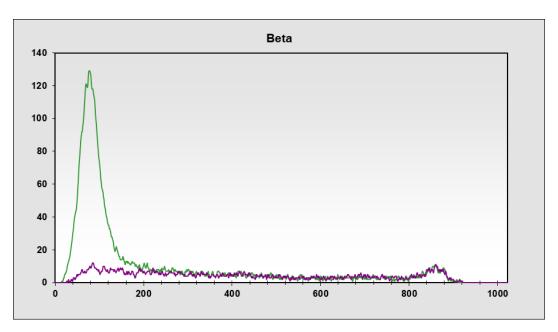


Figure 6. Two separate spectra of vial 35P2 where the sporadic peak appeared and disappeared between channels 0-200.

As stated earlier, these sporadic peaks, only appeared on plastic vials and not glass vials. In addition, these peaks were not present during Part I of the experiment. It was noted that during Part I of the experiment, the humidity was significantly higher in the Fort Collins area. Although the exact value of the humidity of the room was not recorded, the refrigeration unit built into the Hidex 300SL was generating large amount of excess water from condensation, yielding approximately one gallon of water every two to three days. During Part II of the experiment, no excess water was generated. Based on these observations and literature research, it was hypothesized that these peaks were the results of static electricity and a small counting experiment was designed to determine the validity of the hypothesis.

The small counting experiment involved two plastic vials which were counted in 5-minute intervals six times in a row. The vials were counted in the following order: Vial 1, 5-minute interval times six, Vial 2, 5-minute interval times six, back to vial one for 10 total repeats. If the observed phenomenon was due to static electricity, it would be possible that the

static electricity would deposit its energy during the first couple of minutes upon placing the vials into the counting chamber. The results of this test are shown in Table 11 and Table 12:

Table 11. Static Electricity Experiment Result for Vial 1

	Vial 1 - 300s Count, Window 0-200									
Counts	Series 1	Series 2	Series 3	Series 4	Series 5	Series 6	Series 7	Series 8	Series 9	Series 10
1 st 5 minutes	196	1021	321	139	91	105	1063	162	552	1888
2 nd 5 minutes	95	113	90	83	83	87	240	120	117	459
3 rd 5 minutes	21	104	74	94	83	98	135	101	105	355
4 th 5 minutes	99	77	92	94	92	70	119	102	95	129
5 th 5 minutes	102	91	82	72	81	80	109	100	96	116
6 th 5 minutes	90	84	91	82	93	96	116	91	87	109

Table 12. Static Electricity Experiment Result for Vial 2

	Vial 2 - 300s Count, Window 0-200									
Counts	Series 1	Series 2	Series 3	Series 4	Series 5	Series 6	Series 7	Series 8	Series 9	Series 10
1 st 5 minutes	27	74	1424	461	178	424	295	146	1073	112
2 nd 5 minutes	95	99	115	94	88	155	165	102	103	88
3 rd 5 minutes	85	80	123	123	85	141	181	88	97	8
4 th 5 minutes	68	80	88	101	87	552	144	94	86	87
5 th 5 minutes	95	70	101	93	82	487	111	83	79	73
6 th 5 minutes	90	100	90	103	67	289	106	98	92	88

The result of the small counting experiment seems to support the hypothesis that static electricity is the culprit for the sporadic peaks, as most of the high counts appeared during the first 15 minutes post movement of the vial with the exception of Vial 2, 6th series. Using the information gathered from this counting experiment, all of the vials in Part II of the experiment

were recounted to remove the sporadic peaks from the samples. The recounts consisted of five separate 30-minute counts, with the anticipation to discard the data from the first 30 minutes of counting. The decision to discard the first 30-minutes of counting was based on the small counting experiment, as the static electricity peak tends to disappear after 15 minutes; the 30 minutes of stabilizing time was a conservative decision to remove static electricity from the sample. The results of the recounts can be found in Table 13 and Table 14.

Table 13. Part II Experiment Raw Data Recount for Plastic Vials After 7 Days

	Part II - Plastic Vials Recount, Optimized Window 21-185						
	Count1	Count 2	Count 3	Count4	Count5		
7-2P1	1176	530	410	383	494		
7-2P2	511	462	433	452	387		
7-2P3	930	468	413	433	407		
7-2P4	631	470	429	417	456		
7-2PB	1281	594	638	436	398		
10P1	546	447	451	464	466		
10P2	675	421	414	426	427		
10P3	421	421	431	431	405		
10P4	417	467	421	450	452		
10PB	962	399	372	411	423		
14P1	781	535	532	482	478		
14P1 14P2	461	535 448	532 443	482	505		
14P3	452	415	396	493	448		
14P4	429	470	445	367	461		
14PB	1127	691	456	404	432		
21P1	762	471	501	464	456		
21P2	516	442	412	396	400		
21P3	618	454	445	463	485		
21P4	730	562	528	530	523		
21PB	385	455	354	390	372		
35P1	509	439	456	424	447		
35P2	429	436	401	396	425		
35P3	1025	550	453	449	426		
35P4	852	400	387	424	412		
35PB	688	442	381	382	393		
56P1	762	549	478	509	505		
56P2	519	454	455	454	458		
56P3	512	472	465	422	424		
56P4	522	454	414	436	474		
56PB	552	415	403	370	383		
9.4D1	642	452	442	424	126		
84P1	642	452	443	434	436		
84P2	482	449	477	464	441		
84P3	600	472	495	447	434		
84P4	443	433	417	411	440		
84PB	424	412	371	381	408		

Table 14. Part II Experiment Raw Data Recount for Glass Vials After 7 Days

	Count1	Count 2	Optimized Window Count 3	Count4	Count5
7-2G1	620	598	611	592	574
7-2G1 7-2G2	628	630	624	629	648
7-2G2 7-2G3	568	590	593	577	579
7-2G3 7-2G4	518	610	647	644	644
7-2G 4 7-2GB	606	622	580	650	607
10G1	630	572	587	582	627
10G2	629	662	630	575	662
10G2	647	656	640	614	652
10G3	621	592	650	626	667
10G4 10GB	615	599	602	609	567
14G1	670	613	695	580	625
14G2	618	603	604	581	620
14G3	638	633	663	615	601
14G4	666	612	653	597	578
14GB	631	579	563	604	567
TIGE	031	317	303	001	307
21G1	621	619	601	609	570
21G2	660	604	582	585	592
21G3	1163	1174	1194	1162	1203
21G4	2138	2180	2179	2128	2074
21GB	628	564	631	588	623
35G1	618	590	600	600	614
35G2	618	642	636	595	627
35G3	648	644	618	607	623
35G4	701	684	668	669	643
35GB	590	590	623	624	620
56G1	615	631	635	604	669
56G2	570	597	580	593	567
56G3	626	657	654	640	649
56G4	626	599	646	618	622
56GB	602	644	592	594	648
84G1	638	595	591	577	608
84G2	610	579	603	600	616
84G3	590	591	596	625	627
84G4	616	600	603	590	673
84GB	616	587	634	578	551

The recounted data for Part II of the experiment show that the increased low-energy peaks only appeared in the first of the five 30-minutes counts for plastic vials. As previously mentioned, these low-energy peaks are not normal characteristics of tritium behavior; therefore,

for Part II of the experiment, only the recounted data, without the first 30 minutes, are used for statistical analysis.

The statistical analysis is used to determine if the samples in question have a statistically significant increase in tritium activity compared to the controls used in the experiment. Since each vial contains different amounts of deionized water from pipetting uncertainties, the results are normalized via the volume of the deionized water. The volume of the water is determined using mass measurements along with the conversion 1 g/cm³ or 1g/ml, neglecting the 0.3% difference in density from 4 degrees Celsius and 22 degrees Celsius. After the data have been normalized to their respective volume, a student t-test is performed to test the null hypothesis that the sample mean is the same as the control mean. The sample mean is calculated by obtaining the weighted average of the samples for the day. For example, for the plastic samples for day 1 (samples 7P1, 7P2, 7P3, and 7P4), the weighted average is calculated using equation 3.1

$$M_{w} = \frac{\sum_{i} w_{i} M_{i}}{\sum_{i} w_{i}}, \tag{3.1}$$

with,

$$w_i = \frac{1}{\sigma_i^2},\tag{3.2}$$

where M_i is the average of the number of counts within a sample, and σ_i is the standard deviation of the counts within that sample (Cember & Johnson, 2009). This formula is applied to samples that had been counted multiple times.

For samples that have been counted only once, such as the initial data, the data are assumed to be Poisson distributed and therefore the mean count of the sample is the sample

count and the standard deviation of the count within the sample is calculated by the square root of the mean count.

The standard deviation of the weighted mean is calculated by

$$\sigma_{Mw} = \sqrt{\frac{1}{w_1 + w_2 + \cdots \cdot w_n}}.$$
(3.3)

The t-test is then conducted by using M_w and σ_{Mw} of the samples and comparing the calculated sample mean (M_w) and standard deviation (σ_{Mw}) to the control mean (μ_B) and control standard deviation (σ_B) . Using these values, the t-score can be evaluated using equation 3.4 (Cember & Johnson, 2009):

$$t = \frac{M_W - \mu_B}{\sqrt{\sigma^2 M_W + \sigma^2}_B}.$$
 (3.4)

In this experiment, when t is positive, the sample counts are greater than the control and when t is negative, the background counts are greater than the sample. The greater the magnitude of the t-score, the bigger the indication that the sample differs from the control.

Part I Sample Statistical Analysis

Recall for Part I of the experiment, two separate data measurements were performed per sample. The first one is the initial measurement, or immediately adding water and LSC cocktail post drying. After seven days, the second measurement is performed. The student t-test scores per day are compiled in Table 15 and Table 16.

Table 15. Part I – Initial Count T-Score Comparison

Part I – Initial Count T-Score Comparison					
Soaking Time (Days)	Plastic T-Score	Glass T-Score			
1	0.2389	0.2103			
2	0.3270	0.6183			
3	0.0088	0.3150			

4	0.2225	0.0848
5	-0.0896	0.2313
6	0.2301	-0.4232
7	-0.0923	-0.5791

Table 16. Part I – After 7 Days Count T-Score Comparison

Part I - After 7	Part I - After 7 Days of Reabsorption Time T-Score Comparison						
Soaking Time (Days)	Plastic T-Score	Glass T-Score					
1	2.2045	0.7388					
2	1.0798*	0.7537					
3	1.5758	-0.3759					
4	1.1543	1.3154					
5	-0.0782	-1.8307					
6	0.6676	-1.8650					
7	2.0793	1.6339					

^{*}Day 2 plastic t-score had an anomaly as the two measurements of 2P1 were the same, resulting a sample standard deviation of zero. For calculation purposes, as the standard deviation approaches zero, the t-score increases and approaches a maximum of 1.0798. Therefore, the day 2 plastic t-score is a maximum value and not an exact t-score from the sample data.

Part II Sample Statistical Analysis

In Part II of the experiment, recall that the initial measurement was omitted as to allow DI water to interact with the vial without the presence of the LSC cocktail. Results are taken after 7 days of soaking. The results in Table 17 are also taken from the recounted data with the first 30-minutes removed to achieve consistent reading of the samples.

Table 17. Part II - T-Score Comparison

Part II - T-Score Comparison (After 7 Days Reabsorption Time)					
Soaking Times (Days)	Plastic T-Score	Glass T-Score			
7	-1.4273	-0.2877			
10	2.1237	1.3205			
14	-0.4551	1.5123			
21	1.5780	4.1050			
35	0.8262	0.2945			
56	2.8180	0.5472			
84	2.8101	0.4136			

Waste Data

The tritiated water used to soak the vials was transferred into clean plastic vials and measured for comparison purposes. The counts are gathered in the optimized window then divided by the absolute yield of the instrument within the optimized window. The absolute yield is determined by counting the NIST standard in the optimized window and applying it in equation 3.5:

Absolute yield =
$$\frac{\text{measurement}}{\text{standard activity}}$$
. (3.5)

Table 18 and Table 19 show the activity per milliliter of the waste from Part I and Part II of the experiment in units of disintegration per second.

Table 18. Activity of the Waste from Plastic Vials

Activity of the Waste from Plastic Vials										
Plastic Bq/ml	D4	D5	D6	D7	D10*	D14	D21	D35	D56	D84
Vial 1	2194	2185	2188	2198	1486	2126	2121	2095	2091	2066
Vial 2	2189	2184	2177	2187	1437	2121	2134	2097	2091	2076
Vial 3	2187	2188	2190	2187	1556	2130	2120	2095	2085	2076
Vial 4	2187	2191	2185	2182	1466	2137	2131	2098	2093	2081
Average	2189	2187	2185	2189	1486	2129	2127	2096	2090	2075

^{*}D10 waste data had a procedural error yielding skewed data

Table 19. Activity of the Waste from Glass Vials

Activity of the Waste from Glass Vials										
Glass Bq/ml	D4	D5	D6	D7	D10*	D14	D21	D35	D56	D84
Vial 1	2181	2191	2181	2186	1534	2163	2166	2134	2147	2116
Vial 2	2182	2187	2181	2185	1379	2162	2167	2138	2126	2109
Vial 3	2182	2179	2182	2187	1576	2159	2162	2139	2128	2119
Vial 4	2184	2176	2182	2180	1725	2158	2166	2132	2122	2116
Average	2182	2183	2181	2184	1554	2160	2165	2136	2131	2115

^{*}D10 waste data had a procedural error yielding skewed data

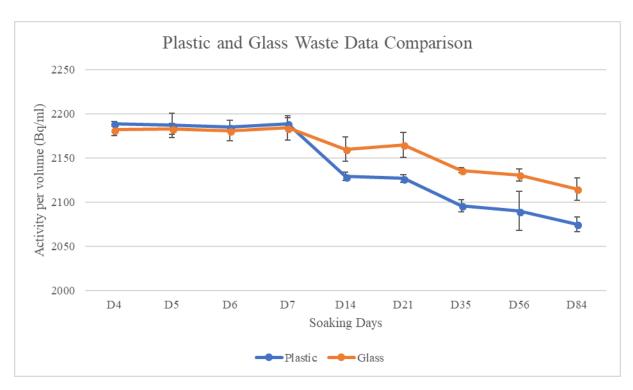


Figure 7. Plastic and Glass Waste Data Comparison. Although there are activity loss per ml of the waste, there seem to have a greater loss of activity in plastic compare to glass. The error bars are based on 2 sigma confidence intervals. D10 waste data had been omitted from this graph due to procedural error.

Tritium Standard Degradation in Plastic LSC Vial Experiment Data

Table 20. Tritium Degradation in Plastic Vial Experiment Data

Tritium Degradation in Plastic Vial Experiment Data								
Vial	S1 = 10	S2 = 14	S3 = 21	S4 = 35	S5 = 56	S6 = 84		
	days	days	days	days	days	days		
Bq/ml	2112	2101	2094	2072	2062	2035		

The results for the tritium degradation in plastic vial experiment in Table 20 can be used to compare to the waste data soaked in plastic in Part II as they utilize the same standard and had undergone the same amount of soaking times. The major difference between the waste data and the aforementioned experiment data is whether the tritiated water had been transferred to a new plastic vial.

CHAPTER 4 DISCUSSION

The t-score is a gauge that indicates whether the samples may differ from the control; the larger the magnitude of the t-score, the stronger the indicator. Strictly looking at the t-score comparison in both experiments, the plastic vials reached a t-score of 2.8 after a soaking time of 56 days. Assuming there is no difference between the sample mean and the background mean, using a degree of freedom of 3, the probability of an individual sample getting a t-value of greater than 2.8 is approximately 0.0339. The probability of having two samples having a t-score over 2.8 out of seven samples is

$$\binom{7}{2} * 0.0339^2 * (1 - 0.0339)^5 = 0.02.$$

Therefore, there is only a 2% chance of obtaining this result, assuming the samples does not contain tritium. A t-score of 2.8 is relatively unlikely to occur by chance within a normal distribution, which potentially indicates that there is an increase in tritium activity in the deionized water. When observing the data in the glass vials, the t-scores remained within -2 and 2, with the exception of day 21 glass vials, even as the soaking time increases. The t-scores between -2 and 2 is approximately 96% of the normal distribution. Although a t-score of 2.8 is unlikely to occur by chance, it is only a subtle increase in activity compared to the control vials.

Comparing the waste results from plastic and glass yielded an interesting observation.

The amount of activity found per milliliter of waste had gradually decreased in both plastic and glass, but the decrease in plastic was greater compared to glass as depicted in Figure 7. The extra decrease in activity in plastic vials seems to be consistent when compared to the tritium degradation in plastic vial experiment. There were three previously mentioned factors that may

contribute to a decrease in activity: degradation of the LSC cocktail, diffusion, and long-term quenching factor. None of these three factors can explain the additional loss of activity within the plastic vials. The LSC cocktail used is the same in both the plastic and glass vials; if degradation had occurred, the changes should be the similar in both plastic and glass vials. In addition, this factor only applies when a sample is stored in the presence of the cocktail and measured before and after storage. For the diffusion factor, since the radiation count was reported in Bq/ml, any diffused tritiated water will be normalized against the volume of the waste provided that composition of the solution is assumed to stay constant. However, both the plastic and glass vials contain the same material, 12 ml of LSC cocktail and approximately 8 ml of standard, the quenching effects should behave the same in both plastic and glass vials. In addition, the TDCR accounts for any quenching effects, negating any long-term quenching changes that may reduce the activity per ml in the waste samples. Furthermore, the long-term quenching factor only matter if the sample is stored in the presence of the cocktail. The loss of activity in the plastic sample in the waste seems to correlate with an increase in t-score as soaking time increases. Between the two observations, it is plausible that some of the lost activity in the plastic vials has reappeared in the deionized water, indicating that there was some tritium exchanged between the plastic vials and the tritiated water. The longer the tritiated water had been soaked, the more atom exchanges could have occurred, and the t-score comparison data seems to support the atom exchange hypothesis. However, the increase in activity of the deionized water seems to be much lower than the activity loss in the plastic vials. It is possible that the deionized water requires more soaking time than 7-days to reach a full equilibrium to allow tritium to exchange back into the deionized water. If the experiment was to be repeated, a longer deionized water reabsorption time should be utilized to attempt to reabsorb the lost tritium in the plastic. It is also possible that

organically bound tritium will be harder to remove which will result in an equilibrium that favors tritium in plastic, rendering a longer reabsorption time ineffective.

The results from the tritium degradation in plastic vial experiment seem to have a slightly lower activity per volume compare to the waste data. This phenomenon can possibly be explained by diffusion. Since the contents of the tritium degradation in plastic vial experiment remained in the vials, it differs from the procedures found in Part I and Part II of the experiment. The mass measurements of the samples after their respective soaking times were not gathered due to an oversight. As a result, the exact volumes of the tritium standard remaining in the vials were uncertain. The volumes used in Table 20 were tritium standard volume data gathered before their respective soaking times during which diffusion could have occurred resulting in a lower activity per volume in the samples.

The glass data observed in day-21 seem to indicate an outlier. Explanations need to be explored and tested to fully understand why two of the vials yielded double and quadruple the counts compared to the glass vials of the same day. The spectra of these two vials can be found in Figure 8, and the two peaks seem to resemble the low tritium standard peaks found in Figure 9. Comparing the heights of the peaks for vials 21G3 and 21G4 to the low tritium standards, it seems that there may be a small tritium contamination at an activity below 2.4 Bq. It is possible that these two vials had a small contamination post the drying procedures which resulted in the increased counts.

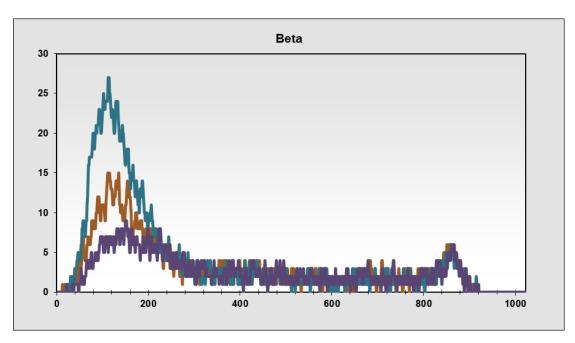


Figure 8. Day 21, glass vials 3, 4 (21G3 and 21G4) and background

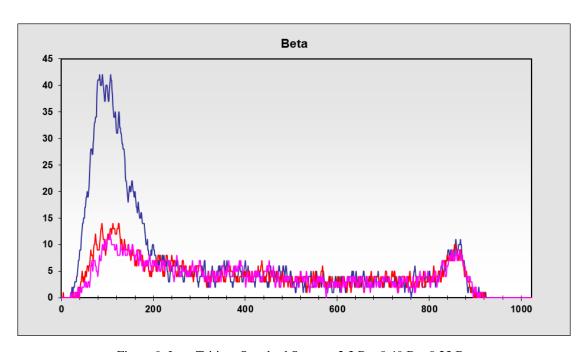


Figure 9. Low Tritium Standard Spectra. 2.3 Bq, 0.49 Bq, 0.23 Bq

CHAPTER 5 CONCLUSION

The experimental results show that within the same timeframe, tritium activity decreases at a faster rate in plastic LSC vials when compared to the glass LSC vials. This result is consistent with the existing research. The excess loss of tritium activity, however, cannot be easily explained as the experimental controls refute some of the current explanations for the phenomenon. In addition, there is some evidence via the student t-tests that suggests that the deionized water used in plastic had traces of tritium in them. These traces of tritium are more evident as the tritiated water soaking time increases, indicating that tritiated water soaking time is a variable that affects tritium reabsorption in deionized water. Together, these two observations support the hypothesis that tritium exchange may have occurred between the tritiated water and the hydrogen within the plastic vials. Although only a fraction of the tritium was reabsorbed from the plastic vials, the atom exchange hypothesis may explain the tritium activity degradation found in plastic vials when the samples have been stored for an extended period of time. Additional research may also be warranted to explore the tritium exchange rate as well as the equilibrium constant to provide appropriate future models for safe storage and usage of tritiated compounds.

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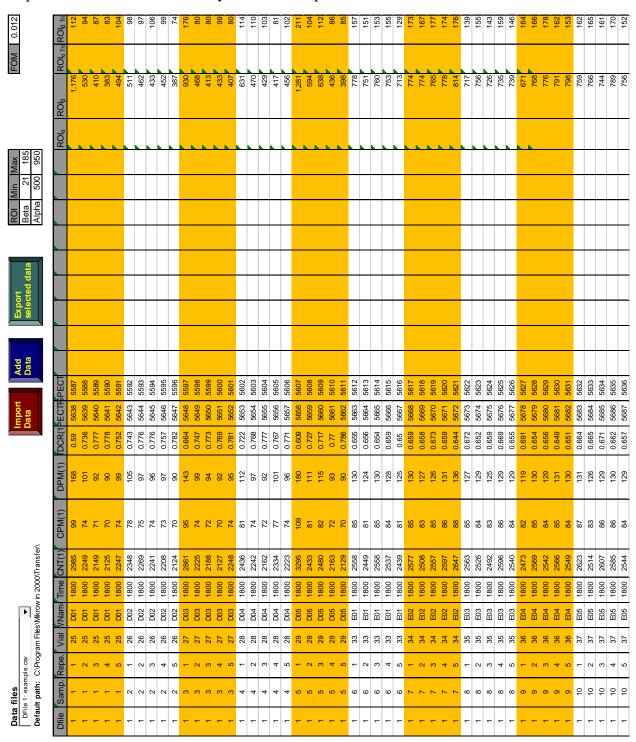
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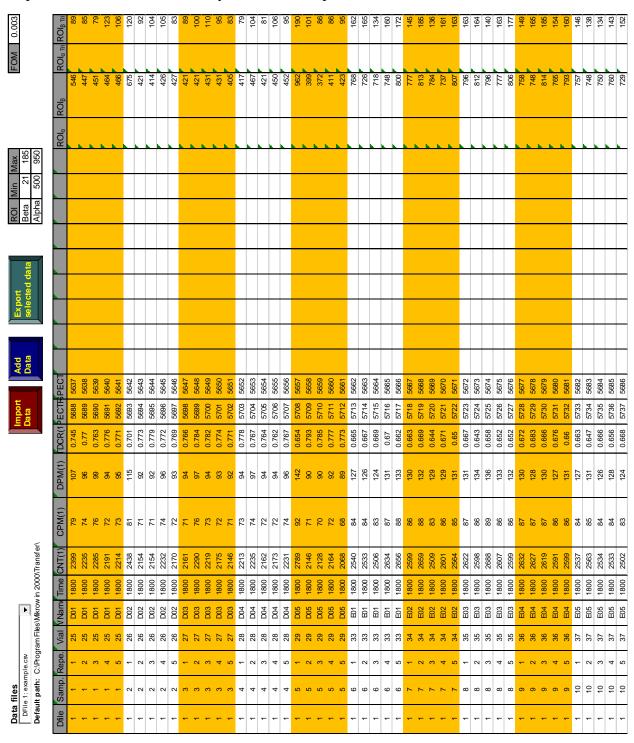
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APPENDIX

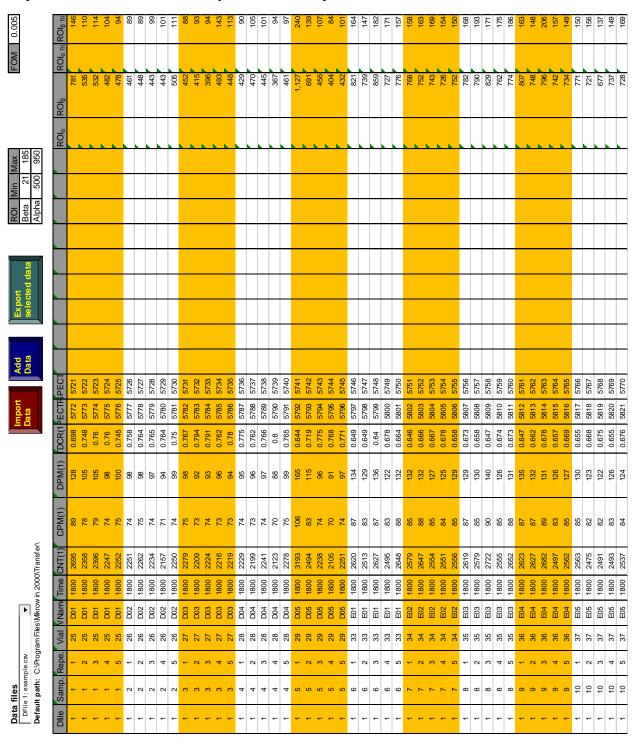
Experiment 2, Plastic Vials – Day 7 Recount, Optimized within windows 21-185



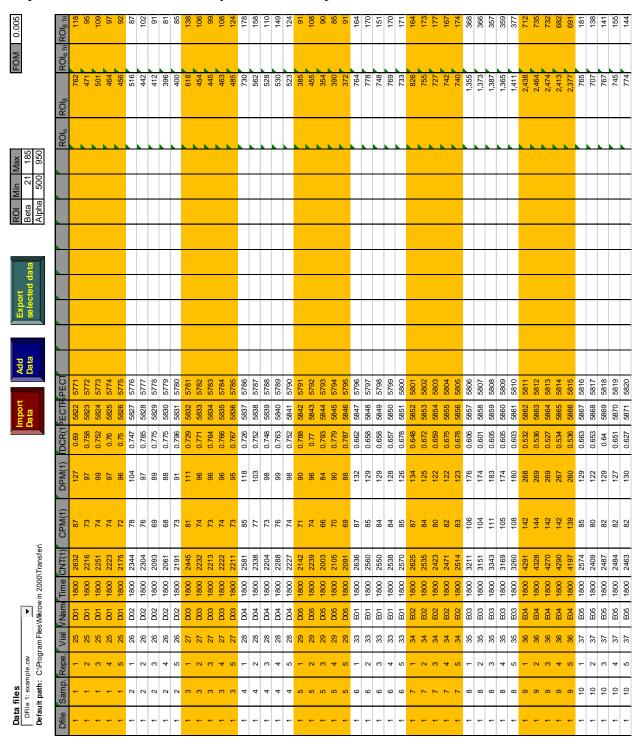
Experiment 2, Plastic Vials – Day 10 Recount, Optimized within windows 21-185



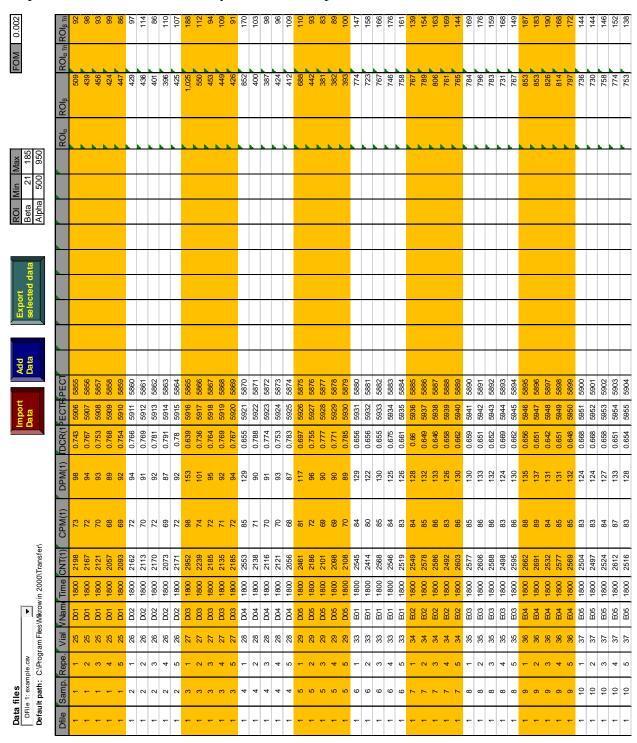
Experiment 2, Plastic Vials – Day 14 Recount, Optimized within windows 21-185



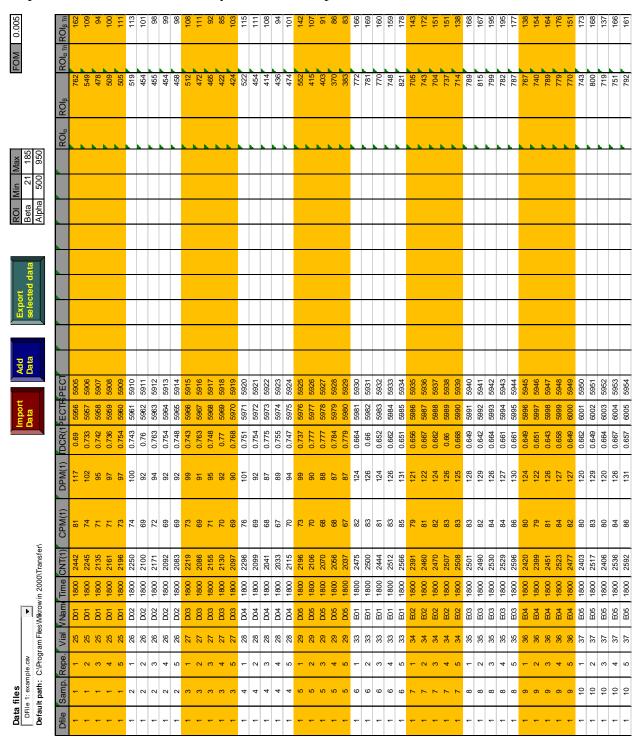
Experiment 2, Plastic Vials – Day 21 Recount, Optimized within windows 21-185



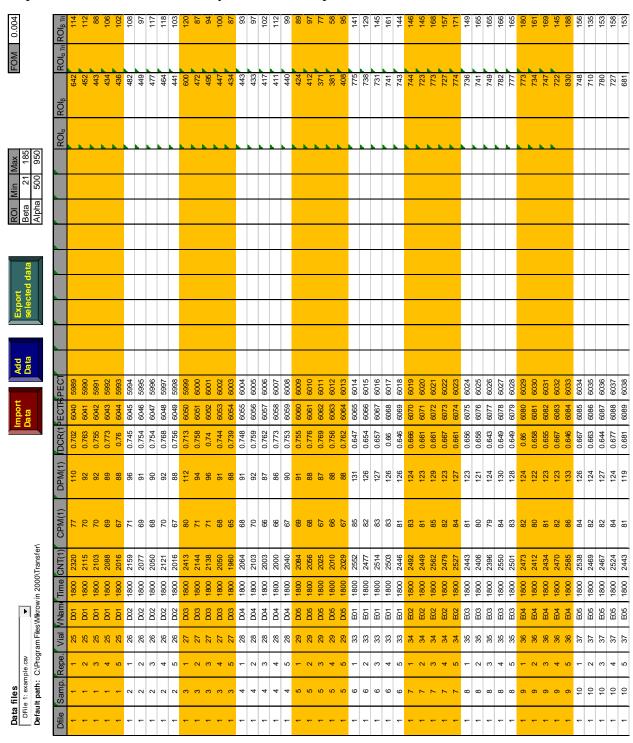
Experiment 2, Plastic Vials – Day 35 Recount, Optimized within windows 21-185



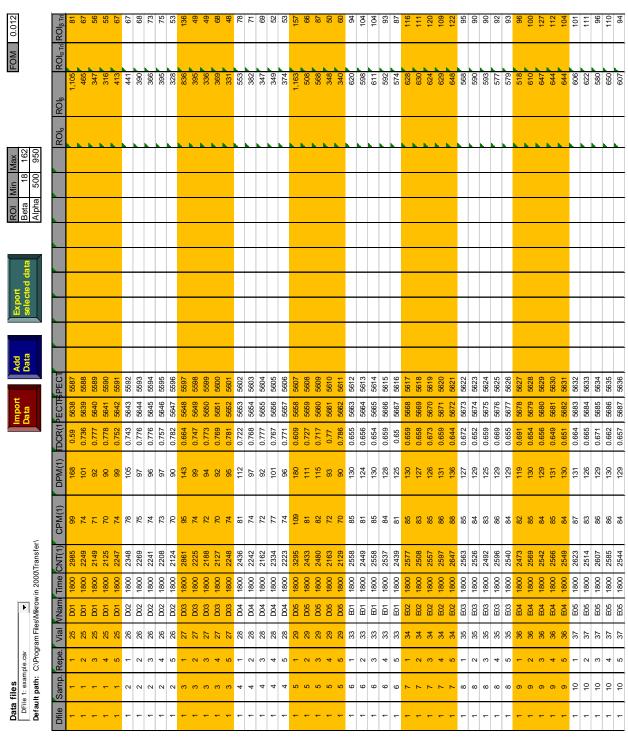
Experiment 2, Plastic Vials – Day 56 Recount, Optimized within windows 21-185



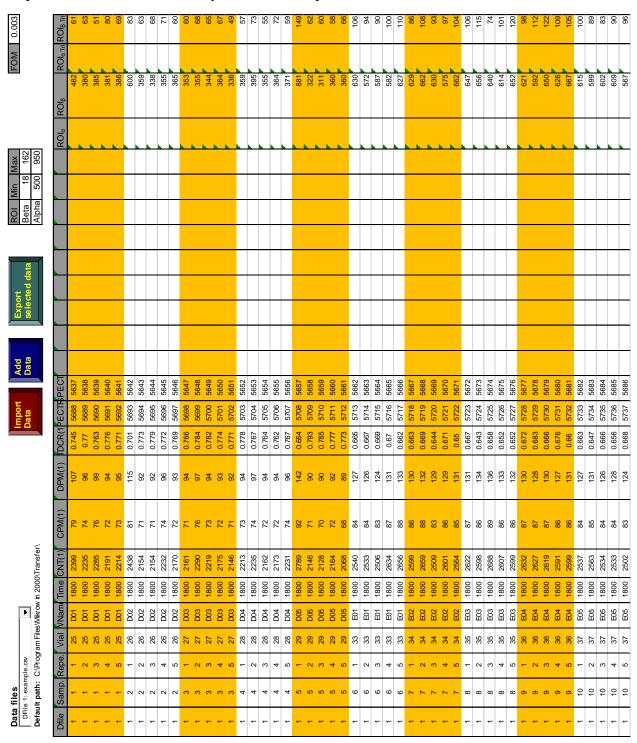
Experiment 2, Plastic Vials – Day 84 Recount, Optimized within windows 21-185



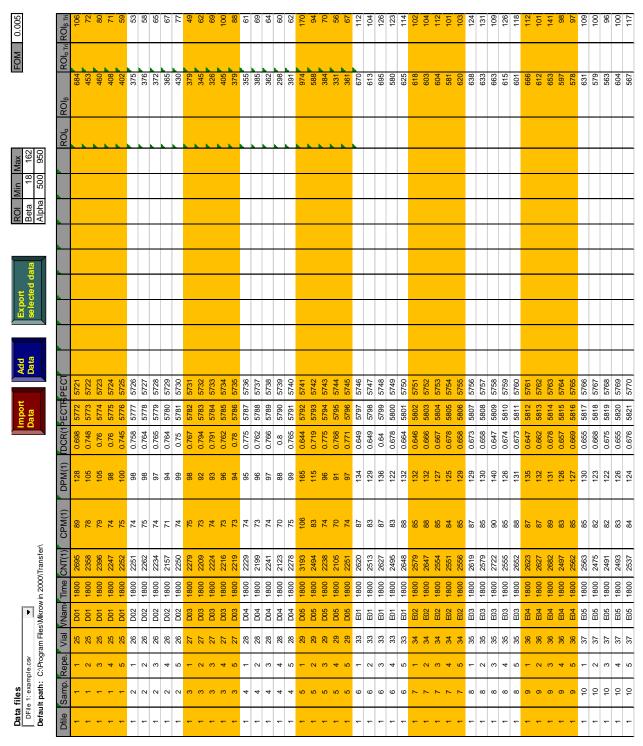
Experiment 2, Glass Vials – Day 7 Recount, Optimized within windows 18-162



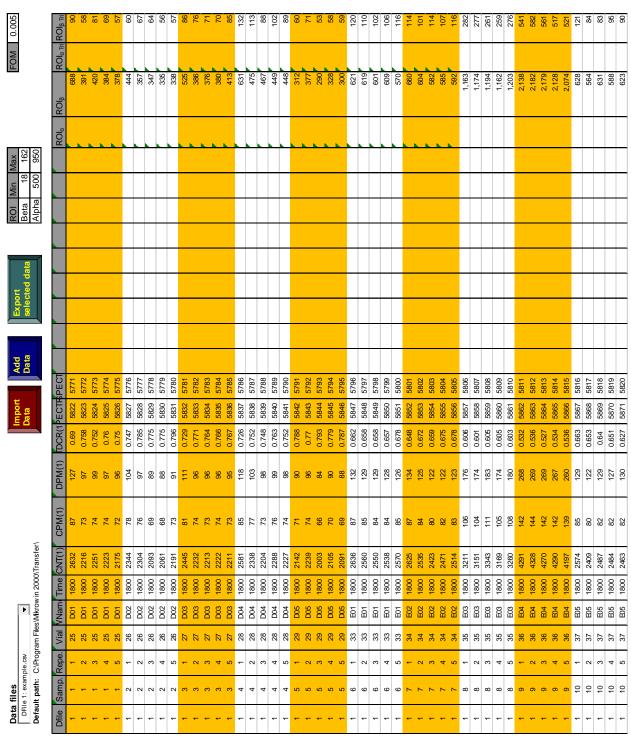
Experiment 2, Glass Vials – Day 10 Recount, Optimized within windows 18-162



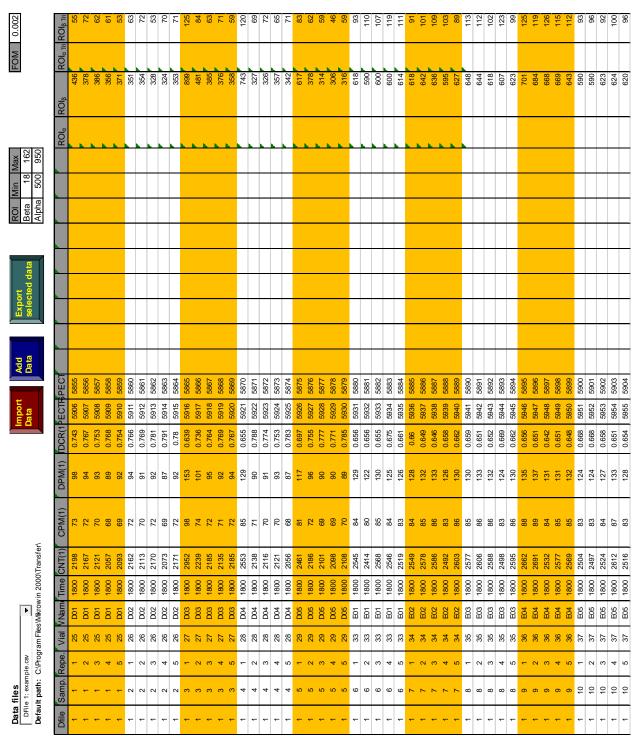
Experiment 2, Glass Vials – Day 14 Recount, Optimized within windows 18-162



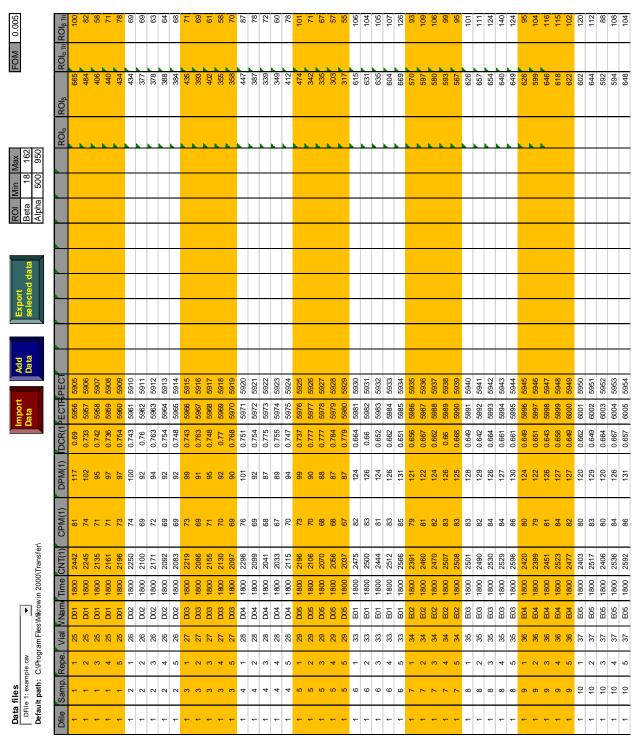
Experiment 2, Glass Vials – Day 21 Recount, Optimized within windows 18-162



Experiment 2, Glass Vials – Day 35 Recount, Optimized within windows 18-162



Experiment 2, Glass Vials – Day 56 Recount, Optimized within windows 18-162



Experiment 2, Glass Vials – Day 84 Recount, Optimized within windows 18-162

