Use Authorization

In presenting this thesis in partial fulfillment of the requirements for an advanced degree at Idaho State University, I agree that the Library shall make it freely available for inspection. I further state that permission for extensive copying of my thesis for scholarly purposes may be granted by the Dean of the Graduate School, Dean of my academic division, or by the University Librarian. It is understood that any copying or publication of this thesis for financial gain shall not be allowed without my written permission.

Signature _____

Date _____

Pressure Optimization in Characterizing Removable Contamination Through Smear

Surveying

By

Joseph Lastor Boring III

A thesis

submitted in partial fulfillment

of the requirements for the degree of

Master of Science in the Department of Nuclear Engineering and Health Physics

Idaho State University

Spring 2022

To the Graduate Faculty

The members of the committee appointed to examine the thesis of JOSEPH L. BORING find it satisfactory and recommend that it be accepted.

Dr. Richard R. Brey

Major Advisor

Dr. Dunzik-Gourgar

Committee Member

Dr. Dewayne Derryberry

Graduate Faculty Representative

Acknowledgements

I would like to express gratitude and acknowledge my committee members, Dr. Mary Lou Dunzik-Gougar and Dr. Dewayne Derryberry, in their support for this research. You have dedicated your lives to the education of future scholars and the betterment of academia. A debt to which we may never recompense save our deepest gratitude. Also, I would like to extend a special thanks to Dr. Richard Brey who has been a tremendous mentor through my academic career at Idaho State University. His commitment to the health physics students of Idaho State University reflects the caliber of excellence he perceives in the Health Physics students.

I would additionally like to acknowledge my family members. I would not be the person I am today without the brilliance of my wife, Laura Hawkins. Her own imperturbable ambition manifests as excellence in those around her and has incited in me enthusiasm to become a better man. She is owed every moment stolen away and given to this piece. My parents taught me that if you give your best effort, then there is nothing more to be asked of you. I have carried that lesson with me and now apply it to every worthy task. Thank you for helping me understand the value of appropriating quality effort before I could learn a more convenient approach. My brothers have and will always be my pride, in all senses of the word. I cannot fathom belonging to another set of gifted men. Lastly, thank you to my work family: Britt Edquist, and Philip Smith. Without your combined insights and charismatic encouragements, my life and my work would be harshly reduced in quality.

Acknowledger	nentsii	ii
Contents	i	V
List of Figures	······································	v
List of Tables		'n
Abstract		ii
1.0 Introduction	n	1
1.1 Back	ground	1
1.2 Proje	ct Justification	3
1.3 Resea	rch Questions	4
1.4 Нуро	thesis Testing	4
2.0 Literature	Review	7
2.1 History.		7
2.2 Compon	ent Definitions1	0
2.3 Swipe C	omposition and Methodology1	2
2.4 United S	tates Regulations	4
2.5 Non-Fix	ed Contamination Analysis1	6
2.6 Remova	l Factor1	9
2.7 Surface	Roughness Categorization2	1
2.8 Minimu	n Detectable Concentration (MDC) and Release	2
2.9 Applicat	ion at Idaho Cleanup Project2	5
3.0 Methods an	nd Materials2	7
4.0 Results and	1 Discussion	3
4.1 Testing	Objective Performance 4	0
5.0 Summary a	and Conclusion	4
6.0 Future Wo	rk 4	5
7.0 References		6
Appendix A.	Removal Fraction Results	7
Appendix B.	R Code Plot Examples	1
Appendix C.	R Code Outputs Examples	2

Contents

List of Figures

3
.8
4
5
6
7
8
9
2

List of Tables

Table 1. 1946 Oak Ridge National Lab Surface Contamination Limitation.	
Table 2. 1947 Oak Ridge National Lab Surface Contamination Limitation.	9
Table 3. 1954 Oak Ridge National Lab Surface Contamination Limitation.	9
Table 4. Swipe Surface Categorization	
Table 5 Removal Fractions for Applied Pressure (Pa) Material Combinations	41
Table 6 Removal Fraction Results	

Pressure Optimization in Characterizing Removable Contamination

Thesis Abstract -- Idaho State University (2022)

This study aims to modify a pervasive method in surveying and characterizing surface contaminated objects, by addressing the ambiguity inherent in common technical procedures. The term "moderate pressure" is an arbitrary statement that is prevalent in technical documentation in reference to performing swipe or removable contamination surveys. This study reviews the literature, introduces an experimental methodology, and quantifies the efficacy associated with the efficacy associated with the application of pressure. The ambiguity surrounding the appropriate amount of applied pressures prompted this study. This paper addresses the optimization of applied swipe pressure when performing removable contamination sampling and quantifies the amount of pressure that should be required and applied pressure of 9,662 to 12,077 pascals onto the swiping medium yielded the optimal removal fraction of contamination for non-porous surfaces, as did 7,246 to 8,454 pascals for porous surfaces, over the equivalent surface area.

Key Words: Health Physics, Pressure Optimization, Smear Sampling, Swipe Survey, Removable Contamination Survey, Removable Radioactive Material Contamination Survey

1.0 Introduction

The objective of this paper was to create efficient and reproducible protocol for conducting smear or removable contamination surveying of a non-fixed radiologically contaminated source. A product of the effort will be to improve the reproducibility of this commonly used sampling procedure. This project was conducted on behalf of the Idaho State University Health Physics program and the Idaho Cleanup Project (ICP).

1.1 Background

The term "moderate pressure" frequently found in the literature, relating to performing smear surveys, is used to describe the amount of force per unit area necessary for an adequate removal of radiological contamination. However, upon review, there is a lack of published research on what the applied pressure may be for smear sample collection when assessing material for free release. Both the Multi-Agency Radiation Survey and Site (MARSSIM) and Multi-Agency Radiation Survey and Assessment of Materials (MARSAME) states that "moderate" pressure should be applied, this terminology is found in other guidance documents such as Nuclear Regulatory (NUREG) Report 1575 and Department of Energy (DOE) Guides. When a person is collecting removable contamination samples using a smear sampling technique, it is difficult to ensure a precise pressure is applied every time without creating a specialized tool. A tool created exclusively for precise pressure of sampling is not economical nor practical and therefore "moderate" pressure with its implied ambiguity has been deemed acceptable. Regardless of the ambiguity of phrasing, the concept of precision or reproducibility is not addressed. Thus, there is a need for defining a quantifiable pressure that is measurable and reproducible in applications such as performing swipe surveys with a robotic arm. With the growing use of robotics in the nuclear industry, this project could lead to a paradigm shift in

removable contamination survey techniques. Defining the pressure of the smear collection for a single swipe to yield a consistent maximum removal without destroying the smear, regardless of surface characteristics such as roughness, is necessary for a robotic smear collection method to produce quantitative and reproducible results.

Defining this pressure was accomplished by setting up an experiment to take removable contamination surveys at a single location multiple times with the same applied pressure, and then measure the quantity of sample matrix removed with each smear. The reproducible applied pressure is repeated with an experimental treatment variable changed, such as the surface material, and this process is repeated until an adequate sample size is established. The sample size was determined by considering the efficacy of an incremental range of applied pressures. If the smear integrity is compromised before a maximum efficiency is obtained, then the smear media may be the limiting factor. Any decomposition of the swiping medium, such as a tear, invalidates a sample. It is important when evaluating performance to understand the effect of variable pressures in order to determine the most reproducible collection efficiency.

There are various ways one could quantify the contamination collected. Two methods of primary consideration are radioanalytical and gravimetric techniques. Though gravimetric analysis is used in this study, it is necessary to understand radioanalytical methods for future applications. The radioanalytical method consists of sample analysis through gamma spectroscopy, alpha spectroscopy, gross alpha/beta proportional counting techniques, liquid scintillation counting etc, as commonly employed. The gross alpha/beta activity in units of disintegrations per minute (dpm)/ 100 cm² or dpm/ 300 cm² will allow comparisons of this data to measurements found in other published field work. Gravimetric analysis takes the pre-swipe weight subtracted from the post swipe weight to determine the remaining net mass. Additionally,

just like radioanalysis, gravimetric analysis has the capability to test materials of various densities and/or particle size.

To make comparisons among different techniques, a sweeping motion covering an area of 100 cm² is desired. A multipurpose robotic arm is thought to be the most useful design, especially if attachments are available to perform different tasks. Since the optimal pressure applied to the swipe is the value that is desired, it is only necessary to measure a pressure applied over a distance. This was accomplished by connecting an attachment, that allows for pressure to be applied to a swipe sample, to a rail that allowed a horizontal movement over a collection surface. This gave the fundamental motion of the test apparatus.

1.2 Project Justification

The scope of the ICP Integrated Waste Treatment Unit (IWTU) includes treating liquid sodium bearing waste in a process that will transform it to a solid-granular material and packaging it in stainless steel canisters to be stored in concrete vaults at the Idaho National Laboratory. A final waste repository does not exist at this time in the United States that is able to accept this type of waste. However, future transport to a permanent disposal site is expected and the waste containers would therefore need to meet the surface contamination levels required by DOT for shipment. Swipe samples will provide the necessary data to determine if containers are in compliance with DOT shipping requirements. Quantification and evaluation of the efficacy and precision of removable contamination survey techniques removes ambiguity of this technique and ultimately it will improve regulatory compliance required in the transportation of waste. Furthermore, use of a robotic arm to perform the swipes potentially ensures ALARA worker dose.

The ICP methods of characterization and decontamination consists of remotely collecting smear samples from high-dose rate waste containers. Due to the high dose rates, these samples will be collected with a robotic arm for evaluation of any present removable material. Within the high dose rate area, a direct measurement of surface contamination will not be possible and therefore cannot be used to verify the effectiveness of the smear sample for collection of a representative removable contamination sample.

1.3 Research Questions

The first set of questions in this research includes (1) "at what pressure for a given surface will the swipe or removable contamination survey be able to most efficiently characterize the contamination source," and (2) "how do factors such as surface types and swipe material impact the results?" It is the goal of both ISU and ICP to establish a method of remotely collecting defensible smear samples from within high-dose rate waste containers, that can be quantified with respect to efficacy and precision.

A potential benefit of this device is the ability to obtain samples from inhospitable locations for characterization. This research provided insight into creating a reproducible swiping method and, it provides data to evaluate another question .(3) "Can a smear sample for removable contamination be used quantitatively instead of qualitatively?"

1.4 Hypothesis Testing

The research questions posed by the apparent ambiguity in literature provides three criteria for test objectives:

1. Reproducibility

$H_{1,0} =$

The relative error of observations in a group is equal to zero indicating results are reproducible with swipe sampling .

 $H_{1,A} =$

The relative error of observations in a group is greater than zero indicating results are not reproducible with swipe sampling.

This evaluates if the technique developed is reproducible. This will be quantified by comparing the observational relative error with the group relative error for each surface and pressure combination. Samples are performed on a uniformly contaminated surface. The relative error for the sample tests where a reproducible method was applied will be assessed using the sample t-test with a cutoff value for determining statistical significance of $\alpha = 0.05$. A p-value of less than or equal to $\alpha = 0.05$ results in rejection of the null hypothesis. A p-value of greater than $\alpha = 0.05$ results in the null hypothesis not being rejected.

2. Pressure Optimization Effectiveness

 $H_{2,0} =$ There is no difference between pressure application for removal fractions. $H_{2,A} =$ There is a difference between pressure application for removal fractions.

The second test objective will be to optimize the amount of pressure applied to collect the greatest mass of representative sample without destroying the sampling media. This will be evaluated by recording the amount of material removed by a swipe sample. This represents the efficacy of sampling the total surface contamination. The sampling environment is not often homogenous nor is the contamination distribution, such as in the case of hot particles. Uniformity of the sampling surface enhances reproducibly, but heterogenous contamination distribution requires direct surveying application and professional judgement for appropriate representation. The population sample mean observed from tests where pressure optimization

was applied, will be assessed using the pairwise sample t-test. A small p-value in a pairwise test indicates a distinguishable fraction of removable contamination between the applied pressures. A large p-value indicates no difference between applied pressure and results in the null hypothesis not being rejected.

3. Quantitative vs Qualitative

 $H_{3.0} =$

There is no difference between experimental removal factor and industry accepted removal factor. $H_{3,A} =$

There is a difference between experimental removal factor and industry accepted removal factor.

The last objective will be to determine if results from swipe sampling may be used as a qualitative or a quantitative measurement. This will be analyzed by comparing the estimated surface contamination values from the swipe sample to the actual surface contamination values, of the test surface contaminated to known levels with the test contaminate. This is a comparison to the industry accepted fraction of removable contamination, 10%, as stated "wipe efficiency may be assumed to be 0.10" (49 CFR 173.443 1992). The population means for the tests, where pressure optimization was applied, will be assessed by calculating the difference between the experimentally measured and industry accepted fraction of removable contamination. The difference is then analyzed using a t-test. A t-test small p-value would indicate any difference of means greater than zero results in the null hypothesis being rejected.

2.0 Literature Review

Non-fixed contamination surveying presents unique issues and may be performed several different ways. While method variability exists, non-fixed contamination is vital to the characterization and understanding of potential health hazards to the workers and the public. The most notable item discussed is the inconsistencies with the United States' regulations concerning non-fixed contamination. This literature review begins with the origin and history of evaluation of surface contamination. The history is followed by definitions of necessary components concerning non-fixed contamination, along with typical sampling methodologies. Additionally, the current regulations of the United States are reported and evaluated. Finally, the review will conclude with context for removable contamination analysis. This literature review will document the importance of non-fixed contamination as a component in the characterization of total contamination.

2.1 History

To understand the importance and impact of sampling non-fixed or removable contamination and its characterization, one should understand the origins of this technique. The sampling media (wipes) used to remove and quantify radioactive contamination were first utilized during the Manhattan Project. It was at Los Alamos National Laboratory (LANL) that a need for characterization of plutonium contamination in the work environment was established. The portable surveying equipment of the time was unable to accurately quantify alpha emitters. Thus, scientists at LANL began using oiled filter-papers to wipe down areas of concern, followed by analyzing the papers with an appropriate alpha counter. (B. C. Hacker 1987) With surface contamination thus appraised, projections of likely airborne contamination levels as a consequence of surface contamination resuspension were the natural next steps in contamination

quantification. The researchers at LANL accomplished this by swabbing the inside of the nostrils of occupational workers with a filter paper, which was then analyzed on an appropriate alpha counter. With the total contamination estimated in this fashion and an understanding of the correlation between surface and airborne contamination, the scientists determined that if the count rate exceeded a 100 counts per minute (cpm) threshold from the nasal swab, then action was required to limit the potential exposures to workers. Oak Ridge National Laboratory (ORNL) took this actionable threshold to another level by requiring that the use of wipes on surfaces such as walls, floors, and benches must also be kept below a 100 cpm level. After World War II and the cessation of the Manhattan Project, the national labs were beginning the formalization of general procedures and appropriate limits on levels of radioactive material as surface contamination. ORNL took the lead in creating a standard methodology for establishing surface contamination levels. Their first implementation of a standard was a 2 in² filter paper wipe, which covered a surface of 12 in^2 . While the initial intention of these wipes was to quantify the amount of plutonium on a surface, ORNL began applying this method to estimate amounts of various other alpha emitting radionuclides on surfaces. These newly standardized limits were established and outlined in Table 1.

Table 1. 1946	6 Oak Ridge	e National 1	Lab Surface	Contaminatio	n Limitation.
	0				

Location	Surface Contamination Limit (dpm)
Exposed Surfaces, Floors, Bench Tops, ETC	0
Exposed but Protected from Handling	25

Inside Poor	25
or	
Intermittently	
Used Fume	
Hood	

After a year of using these limitations, ORNL decided to expand this approach to distinguish radionuclides. This led to the incorporation of limits placed on beta and gamma emitting contaminations. Thus, the tolerance levels for surface contamination needed to be upgraded, as shown in Table 2.

Table 2. 1947 Oak Ridge National Lab Surface Contamination Limitation.

Location	Surface Contamination Limit
Tabletops, Floors, and Apparatus	0 dpm Alpha, 200 dpm Beta/ Gamma
Boxes for Shipment by Air or Rail	0 dpm Alpha, 0 dpm Beta/ Gamma

It was not until around 1954 that ORNL established limitations on surface contaminants similar to the standards used by the United States currently. With this update, Oak Ridge recommended that the wiping method must be conducted over 100 cm², or roughly the size of a single dollar bill. Additionally, the national lab began to use a general surface tolerance, instead of addressing each surface individually. This standard is highlighted in Table 3.

Table 3. 1954 Oak Ridge National Lab Surface Contamination Limitation.

Location Surface Contamination Limit

General	30 dpm Alpha,
Power	1000 dpm Beta/
Plant	Gamma
Surface	

While ORNL and LANL attempted to establish recommendations for removable surface contaminants, the rest of the United States' nuclear industry did not institute this approach due presumably to its questionable quantitative nature. During this time period, the main use of this wipe method was to determine whether or not there is a contaminant, not necessarily how much or what constitutes the contaminant. This is seen within a quote from (National Bureau of Standards Handbook 51. 1952.), "Estimates made by counting wipes furnish useful information and should be made in order to determine whether or not a contaminant is likely to rub off a surface. However, these measurements are only qualitative and unreliable because of the uncertainty as to the amount of contamination removed." This excerpt reflects a concept that was constant during the early developmental period of the wipe method sampling of removable surface contamination and that is still observed, as discussed in section 2.4 United States Regulations.

2.2 Component Definitions

Beside the ambiguity in precision, there are other inconsistencies with this approach. Broadly speaking, the term surface contamination may be broken into two major components: fixed and non-fixed. This review does not discuss fixed contamination, instead it focuses on the non-fixed removable contamination. The first concept to clarify is the precise definition for a non-fixed contaminant. Unfortunately, this term is not universally defined, and it varies in characteristics depending on the regulator. However, non-fixed surface contaminants may be described as removable, transferable, smearable (meaning a surface contaminant that is readily sampled), or a loose contaminant. While regulatory agencies may debate its precise definition, a non-fixed surface contaminant may be fundamentally characterized as a contaminant that is capable of having a fraction of its total readily removed from a surface. This leads to a second challenge; the necessity in defining surface contamination. However, it is initially prudent to recognize the term total contamination, as the summation of fixed and non-fixed contamination. This is a concept that leads to the characterization of total contamination. This summation is typically quantified through Geiger-Mueller detector sweeps of the surface to evaluate count rate, or the use of stationary analytical equipment to evaluate contamination levels. DOE regulation (10 CFR 835 Appendix D) states that "It is not necessary to use swiping techniques to measure removable contamination levels if direct scan surveys indicate that the total residual surface contamination levels are within the limits for removable contamination".

While the threshold limitations for wipes have been discussed, the description of a wipe has yet to be set. A wipe is the medium used to sample a contaminant by removing it from a surface, this may also be referred to as a smear, swipe, or swab.

Direct scan surveying methods themselves are often hard to assess. As an example, when LANL was attempting to quantify the amount of plutonium in the workplace, they reported that an accurate representation of the radionuclide was not possible through direct scan surveying methods. Plutonium's characterization could be accomplished only through swiping techniques and laboratory analysis these approaches allowed for the analysis of Plutonium in samples. The ability to identify the various alpha emitting radionuclide contaminants on swipe samples and the eventual ability to identify unknown beta and gamma emitting radionuclides led to the wipe sampling techniques becoming more prevalent. Nevertheless, surveying limitations of wipe quantification is evident.

2.3 Swipe Composition and Methodology

Professional judgement and field experience are the primary basis upon which the precise locations for evaluating removable contamination are chosen. Professional judgement of swipe surveying identifies locations of primary concern for potentially spreading contamination and these subsequently require characterization. Some common locations to be considered include: Places where unsealed radioactive material was handled or stored. Or areas where contamination is likely to be spread to by humans such as: phone handles, door handles, refrigerator handles, light switches, sinks, floors near doorways, eating areas, areas of frequent foot traffic etc. If the surface contamination is generated by the deposition of aerosols from the air, the contaminant is likely to have a uniform distribution. Additionally, the greater the quantity and toxicity of the radionuclide employed, or more crucial the consequences of removable contamination, the more frequently evaluations of removable contamination should be performed.

With the locations of surveying in mind, the regulatory agencies have elected to move towards nuclide-specific dose-based limits for total surface contamination rather than somewhat arbitrary limits on concentration of radioactive material measured on removable contamination surveys. "The results of smear [removable contamination] samples should not be used for determining compliance. Rather, they should be used as a diagnostic tool to determine if further investigation is necessary." (MARSSIM 1997) Wipe tests to assess the concentration of removable contamination may be used to provide qualitative indications of removable activity and satisfy (As Low as Reasonably Achievable) ALARA requirements.

The following recent guidance from the Health Physics Society's ANSI N13.4 standard states that:

- Removable contamination surveys should be performed by using smears, cottontipped swabs, and/or chemically treated cloths.
- A wetting agent may be used to increase collection efficiency.
- Large-area smears surveys may be used to supplement standard smear techniques in areas generally assumed not to be contaminated, such as control points.
- An area of approximately 100 cm² should be smeared.
- An S-shaped survey should be of approximately 40 cm in length.
- The smear should be pressed moderately against the surface to be assessed.
- Smears should be individually numbered.
- Smears should be handled in such a manner as to prevent cross-contamination.

Guidance in the Department of Energy regulations (10 CFR 835 Appendix D, 1998) states: "The amount of removable activity per 100 cm² of surface area should be determined by swiping the area with dry filter or soft absorbent paper, applying moderate pressure, and then assessing the amount of radioactive material on the swipe with an appropriate instrument of known efficiency. (Note: The use of dry material may not be appropriate for tritium)." However, what constitutes moderate pressure is not defined.

Swiping techniques may be taken in many patterns: an inwardly spiraling circle, a straight line, zig-zagging area, etc. The precise pattern is not important, but the method and material for collecting the swipe sample should be standardized as much as possible, to create reproducible data. A wide variety of types and sizes of wipes are used to collect non-fixed contamination: circular disks and squares made of cellulose, fiberglass, cloth, or plastic; cotton swabs are also used. (ISO-7503-1 1988) recommends that "the smear material should be chosen to suit the surface to be checked (for example filter paper for smooth surfaces, cotton textile for rough surfaces)."

The guidance regarding the area over which smears should be taken and averaged is somewhat inconsistent. The standard area for a single wipe is 100 cm², but 300 cm² is sometimes permitted. Department of Energy Regulations (10 CFR 835 App. D, 1998) specify the total (fixed plus removable) surface contamination can be averaged over 1 m². NRC Regulatory Guide 8.21 agrees by stating total surface contamination is "acceptable to average removable contamination on objects in restricted areas over 300 cm², but contamination on floors, walls and ceilings should be averaged over 100 cm²". This is consistent with DOT regulations specifying that individual smears on packages offered for transport can be performed over 300 cm². Depending on the roughness of the surface and the type of wipe material, wiping over too large an area can destroy the physical integrity of the wipe material. Limiting the area being evaluated to 100 cm² is in most cases described as appropriate to avoid degradation, according to NRC Regulatory Guide 8.21, and 8.23.

2.4 United States Regulations

The United States' regulations pertaining to swipe tests are not consistent as to the question of if wet or dry swiping medium should be employed. 10 CFR 835 Appendix D for surface contamination values notes that: "the amount of removable radioactive material per 100 cm² of surface area should be determined by swiping the area with dry filter or soft absorbent paper, applying moderate pressure, and then assessing the amount of radioactive material on the swipe with an appropriate instrument of known efficiency." An exception to this approach is suggested for evaluating removable tritium contamination. When sampling for removable tritium contamination "a standard smear measurement [can be used,] but using a wet swipe or piece of Styrofoam" (DOE Memorandum 1995) is suggested. Tritium is a special case, in part because there is no satisfactory way to measure total tritium surface contamination with a survey meter.

Since it is also prone to penetrate the surface, tritium is often missed with a dry smear. Conversely, the use of a wetting agent may lead to a significant underestimation of the contamination in the case of alpha-emitters due to self-attenuation. The potential for significant attenuation of low energy beta emitters or alpha radiation when using some analytical protocol such as proportional counters should also be considered when determining the appropriate smear collection technique.

It is not always obvious whether it is necessary to calculate the level of non-fixed contamination collected on a single wipe or to estimate the total amount of non-fixed contamination on a surface. As one can not collect all of the material on the surface, it is logical to refer to estimating the surface concentration. If the regulation refers to a "wipe" limit, it applies to the contamination on a single smear. If the limit is simply referred to as a contamination limit, it could apply to either the contamination on a single smear or the total amount of removable contamination on a surface. The assumed efficiency of sample collection is seldom mentioned.

Limits on total non-fixed contamination cannot be adjusted. However, wipe limits (pertaining to contamination on a single wipe) that assume a removal efficiency of 10% (49 CFR 173.443 1992) may be adjusted. It is entirely reasonable to adjust these limits when the wipe sampling efficiency is known to be different than this default assumption. To determine if the removal fraction was greater than the default assumption of 10%, it may be determined experimentally ("exhaustive removal by repetitive smears"). If it was 20% (for example and well documented for a particular situation), the non-fixed contamination limit is doubled. The adjustment to the limit is made in the following manner from US DOT's 49 CFR 173.443:

$$L_{adi} = L_{0.1} * \frac{F}{0.1}$$
 (Equation 1)

Where:

- L_{adi} = adjusted wipe limit, the maximum value of which is ten times the unadjusted limit.
- $L_{0.1}$ = unadjusted wipe limit that assumed a removal fraction (collection efficiency) of 0.1; and
- F = estimated removal fraction of the wipe, the maximum of which is 1.0.

2.5 Non-Fixed Contamination Analysis

A key issue is the interpretation of swipe measurements with no removable contamination detected. Without a statement of the measurable level of removable activity, such a result contains little useful information. The minimum detectable activity (MDA) quantifies the detection capability of an analysis technique nested in a statistical confidence interval to provide a probability of detection.

There are two ways to measure surface contamination: directly and indirectly. Direct measurements of surface contamination are made by means of a field survey meter or monitor. A direct measurement estimates the combined fixed-plus-removable contamination but may also include contributions from interfering radioactivity inherently present in the surface or from ambient sources in the area adjacent to the measurement. An indirect evaluation of removable surface contamination is made by taking and subsequently analyzing a swipe sample or a grab sample.

If removable contamination levels are present at low levels, they may not be detectable by use of swipes due to poor swipe collection efficiency. Additionally, such low levels may not be detectable with portable survey instruments where a direct surface measurement is made, due to interference from background activity, source to detector geometry, or detection instrument efficiency. The only technique for assessing uniform contamination at low levels with a greater degree of certainty was by use of "grab" sampling of the material with subsequent destructive analysis at a laboratory. A grab sample is like a snapshot in time of any individual sample collected without compositing or adding other samples. Thus, the use of swipes, field monitoring equipment, and grab sampling for laboratory analysis are all important components of assessing the state of radioactive materials contamination of a surface. Field measurements and grab samples generally will result in an estimate of the sum of fixed and removable activity, whereas swipes generally will result only in an estimate of removable activity.

The level of non-fixed contamination on a smear is sometimes determined with a survey instrument in the field before the wipe is transported to the laboratory. "Wipes should not be left uncovered for extended periods prior to counting, since this increases the possibility of radon decay products accumulating on the smears [or for volatile materials to escape]. Wipes can pick up radon decay products from the surfaces being tested." (Frame 1999)

Liquid scintillation analyses are the most used technique for analyzing low energy beta emitters (e.g., H-3, C-14, S-35). Gamma scintillators (often well detectors) may be used for quantifying radionuclides that do not emit charged particles. Otherwise, a proportional counter, Geiger Mueller (GM) detector, alpha scintillator or beta scintillator may be used. When a swipe sample is analyzed by liquid scintillation counting (LSC), it is common to cut up, roll, or otherwise fold the smear and place it in the liquid scintillation vial. Unfortunately, the counting efficiency can be low due to the absorption of light by the smear sampling media or the absorption of the emitted radiation by the wipe matrix, unless translucent materials transparent to the wavelength of light providing the highest PM-Tube photocathode quantum efficiency are used. Wipes should be used that become translucent when immersed in the liquid scintillation cocktail (e.g., glass fiber). "When counting smears by LSC, it is not uncommon to encounter

chemiluminescence, as a frequent source of this chemiluminescence is the accumulation of cleansers and chemicals on the wipe." (Sturchio 1999) Chemiluminescence is the emission of light as the result of a chemical reaction and may give the appearance of tritium with an increase in count rate. Whether or not the emitted light is due to chemiluminescence or the actual presence of tritium can be evaluated by performing multiple counts and observing a cessation of chemical reactions. LSC is a particularly effective method for analyzing wet smears. If a wet smear were counted with a standard counting system (e.g., GM, proportional counter, alpha or beta scintillator), the moisture in the smear matrix could result in counting losses due to self-absorption of alpha or low energy beta particles. If this is the case, once the swipe sample has been counted, the following equation (Frame and Abelquist 1999) for calculating the total removable surface contamination, as (dpm/cm²) can be used:

$$A_s = \frac{R_{s+b} - R_b}{\varepsilon_i * \varepsilon_s * F * S}$$
(Equation 2)

Where:

 A_s = total removable surface contamination.

 R_{s+b} = gross count rate on the smear counter.

 R_b = background count rate.

 ε_i = instrument efficiency, i.e., the fraction of the particles leaving the surface that produce a count.

 ε_s = source or surface efficiency, i.e., the fraction of the decays within the sample that result in a particle (quantum) of radiation leaving the surface of the source. F = removal fraction; and

S = surface area covered by the smear, e.g., 100 cm².

Non-fixed contamination on surfaces can result in personal exposures via external

exposure to gamma rays, or higher energy beta particles, inhalation of resuspended airborne

contamination, ingestion of contamination picked up on the hands, and external exposure due to

submersion in air borne contamination. Therefore, if the level on non-fixed surface

contamination must be evaluated to determine compliance with specific limits, it must be known

whether these limits refer to the total removable contamination on the surface or to the contamination picked up on the wipe sample medium. Frame and Abelquist (1999) observe that, "At a minimum, it may be concluded that removable activity is present when smear [swipe sample] results exceed the critical level of the smear [swipe sample] analysis procedure. If a smear [swipe sample] result indicates that activity does not exceed background levels, it might be inappropriate to conclude that removable activity is not present since the collection efficiency could be near zero." If a swipe sample shows contamination, it confirms that there is removable contamination present. However, if a swipe sample does not show contamination, it cannot automatically be concluded there is no removable contamination present. Note that directly measuring the surface using a field instrument will give an estimate of the sum of the fixed-plus removable activity.

2.6 Removal Factor

The removal factor is the ratio of the activity of the radionuclides removed from the surface by one swipe sample to the total removable activity of the surface prior to this sampling. The removal factor, F, is defined by the following relationship:

$$F = \frac{A_l}{A_{sum}}$$
 Equation (3)

$$A_{sum} = \sum_{i} A_{i}$$
 Equation (4)

Where:

 A_l = activity removed by the initial swipe sample. A_i = activity removed by the ith swipe sample; and A_{sum} = total activity of the removable surface contamination prior to taking the first swipe sample; estimated by summing a set of repetitive swipes.

"The removal factor should be determined experimentally for each set of measurement conditions encountered, using the method of exhaustive removal by repetitive swipe sampling over the same surface area." (EPA 2011) The sum of the activities that are removed by repetitive swipes yields an estimate of the total removable activity A_{Sum} . This can then be compared to the activity removed by the initial swipe sample, A_l , to yield the removal factor. If it is not possible to determine the removal factor experimentally, a value of F = 0.1 (i.e. 10%) is sometimes assumed (see for example 49 CFR 173.443(a)(1)). Depending on what, if any, prior knowledge of the surface characteristics are available, the use of this 0.1 value as a default may introduce uncertainty into the result that may not be reflected in the uncertainty reported. The 0.1 value is chosen to represent a conservative 10% approximation of the removal factor from the initial swipe sample.

Under the influence of natural physical and chemical interactions in the environment, fixed contamination may become removable, or removable contamination may become fixed. If the removal efficiency is very low and the first swipe is the only one that shows significant activity, the removal factor may be erroneously overestimated as 100%.

It seems to be universally accepted, although not commonly acknowledged in practice, that only a "fraction of the [removable contamination] is sampled with the first swipe." (EPA 2011) Some of the parameters that affect the removal factor are the chemical form of the contaminant and how the contaminant interacts or is absorbed on the surface under consideration. This can affect the fixed component, but the chemical form of the contamination will also affect the removal factor, depending on whether dry or wet swipes are used.

The removal factor will depend on whether the contaminant has a greater affinity for the surface being sampled or the swipe material. Jung et al. (2001) found it took up to 10 consecutive swipes to obtain a good estimate of the total removable activity, even on relatively smooth surfaces, with the initial swipe having a removal factor of 10% to 20%. The variability in

swipe results, even from surfaces expected to give consistent results, was greater by an order of magnitude or more than the counting statistical errors. The document ISO 11929-7 (2005) contains an example calculation for the uncertainty and limit of detection for swipe samples. This calculation models the removal factor with a rectangular distribution from 0.06 to 0.62 with a mean of 0.34.

Yu et al. (2003) reviewed the literature and concluded that a removable fraction of 10% was appropriate, by modeling the removable fraction as a triangular distribution from 0 to 1 with a mode (most likely value) of area XYZ. The removal factor can be determined experimentally using the ISO 7503 method of "exhaustive removal by repetitive [s]wipe tests." The step-by-step addition of the removable activities leads to an approximation of the total removable activity (A_{sum}) to which the activity removed by the initial swipe removable contamination test (A₁) can then be related to the yield of the removal factor.

2.7 Surface Roughness Categorization

A review was conducted to evaluate the swipe surfaces and how they might be categorized by surface roughness of the material swiped. Classification of different surface roughness will allow a more appropriate determination of the amount of removable surface contamination transferred to a swipe sample. "Swipe surfaces fall into four categories: rough non-porous, rough porous, smooth porous, and smooth non-porous." (EPA 2011) Table 4. "Swipe Surface Categorization" lists the swipe surfaces used in this study and how they would be categorized according the EPA recommendations.

Table 4. Swipe Surface Categorization

Category Swipe Surface

Rough	Unpainted
Porous	Concrete
Rough	Painted
Non-	Concrete
Porous	
Smooth	Second
Porous	Layer
Smooth	Linoleum
Non-	Stainless
Porous	Steel

2.8 Minimum Detectable Concentration (MDC) and Release

MDC values are primarily used with laboratory instruments utilizing a counter for measuring total counts in a specified time while holding the geometry constant. Both the scan and pausing for investigating an increase in counts when using a portable handheld detector arrangement are considered qualitative measurements, whereas laboratory measurements are considered quantitative measurements albeit with substantial sampling variation. Such laboratory measurements may be performed to quantify a count increase confirmed during the pause required as part of the survey plan. Measurements are performed with a 60-second duration in proximity or in contact with the source of contamination; however, this may vary depending on the Data Quality Objective requirements. MDCs are based on a Poisson approximation of the backgrounds standard deviation.

MARSAME (2009) states that the static minimum detectable net signal (S_D) should be based on a Type I and II error (α – value and β – value) of 0.05 to minimize false positive and false negative occurrences.



Figure 1. Probability of Detection

Figure 1 (MARSAME Figure 7.8) illustrates a sample distribution representative of background with the mean specific to Natural Background (N_B), and a sample distribution above background when residual radioactivity is present (with the mean specific to the S_D) with a possible Type II error compared to the critical value of the net instrument signal (S_C). The Type I and Type II errors were assumed to be 0.05, indicating that there was a 5% probability of a sample distribution with no residual radioactivity at the $S_{\rm C}$ threshold; whereas the Type II error indicated that there was a 5% chance of a sample that actually contained residual radioactivity being considered part of the background distribution at the $S_{\rm C}$ threshold. Measurements that exceed the S_C threshold, while not exceeding the S_D threshold, may need to be investigated further by taking more measurements or increasing the count time where the average reading may be used for comparison to the action level. The action level as defined by MARSSIM may be a regulatory threshold standard, a dose or risk based concentration level, or a reference based standard. The action level comparison can also be considered an investigation level that is a derived media-specific, radionuclide-specific concentration or activity level of radioactivity that: 1) is based on the release criterion, and 2) triggers a response, such as further investigation or cleanup, if exceeded. If residual radioactivity is present, then the average of the measurements

(or the single measurement if the S_D threshold was exceeded) should be compared to the action level to determine if the item may be released to the public.

According to MARSAME there are two initial decisions made when evaluating material for release. These decisions determine if the material is considered impacted or non-impacted. This assessment involves considering process knowledge. Non-impacted material is material with no reasonable potential to exceed background radioactivity levels; whereas, impacted material has a potential to exceed background radioactivity levels. Material determined to be "non-impacted" do not require additional surveys or evaluations to release the material. Survey measurements may be performed to confirm whether the material is non-impacted. Whereas, impacted material requires further evaluation, which consists of additional process knowledge, visual inspection of the material and/or performing radiological surveys. There are three classifications of impacted material (MARSAME 2009):

- Class 1 Materials and Equipment (M&E) are those that may have been in direct contact with radioactive materials during operations or may have become activated and are likely to exceed the action level.
- Class 2 M&E are impacted M&E that have, or had, (1) low potential for radionuclide concentration(s) or radioactivity above the action level(s); and (2) little or no potential for small areas of elevated radionuclide concentration(s) or radioactivity. Such potential may be based on historical information, process knowledge, or preliminary surveys.
- Class 3 M&E are impacted M&E that have, or had, (1) little, or no, potential for radionuclide concentration(s) or radioactivity above background; and (2) insufficient evidence to support categorization as non-impacted. In some cases, static measurements should be performed when there is concern that the scan MDC would exceed the action level, and are recommended for alpha contamination surveys when comparing to the transuranic authorized limit of 100 dpm/100 cm².

2.9 Application at Idaho Cleanup Project

The IWTU facility will treat liquid sodium bearing waste in a process that will transform waste to a solid-granular material, and package it in stainless steel canisters to be stored in concrete vaults at the Idaho National Laboratory. To meet DOT and waste acceptance criteria for the final disposition site, a high degree of cleanliness is required. However, during various operational start-up activities and equipment testing, it was determined that the existing ability to maintain the exterior of the canister free from radioactive contamination or recovery from an operational upset condition is limited. Currently, once the canister is loaded and sealed it is moved away from the fill head area where a pair of manually operated tele-manipulators are used to survey the exterior of the canister for removable contamination. Should removable contamination be present on the canister, the tele-manipulators may be used with an existing vacuum wand or specialized swabs to further clean the canister. However, once radiological operations commence, entry into the canister fill cell will be severely restricted. Failure to effectively clean the canister initially would restrict withdrawal of the canister from the canister fill cell, interrupting or shutting down waste processing. The DOT requirements and RH-72B transportation cask requirements, limit removable beta-gamma surface contamination to 200 dpm per 100 cm² and removable alpha surface contamination to 20 dpm per 100 cm².

To prevent contamination greater than regulatory limits, two facility modifications for IWTU were implemented. The canister fill carts were modified to accommodate a new position and a robot arm was added in the canister fill cells to support canister decontamination in the operations position. The robotic arm can be used to check for contamination and decontaminate a canister as necessary to allow for removal from a canister fill cell. The Canister Decontamination System (CDS) uses the robotic arm to perform multiple operations on the canister which includes

swipe sampling for removable radioactive contamination, vacuuming, and manual decontamination. Additionally, the in-cell master slave manipulators will support the robot by transferring robot end effectors to and from the glove box.

Additional improvements may be required to ensure IWTU product canisters can be efficiently checked for contamination and decontaminated as necessary, immediately after the canister filling operation. Additional position sensors may be required to ensure the cart and robot arm are coordinated. Another improvement is the quantification of pressure the robotic arm must apply to the canister for proper characterization (using swipe surveys) and for decontamination purposes. Presently the robotic arm surveys specific locations as the cannister turns in place on a cart system. Distinguishing the appropriate applied pressures exerted by the robotic arm during survey operations will allow for the cleanliness of the cannister to be upheld and proper removable contamination characterization accomplished for eventual transportation.

3.0 Methods and Materials

The primary focus of investigation in this research were the affect and optimization of pressure, and also combinations of the swipe material, and surface materials that produced the most effective sampling efficiency. The controlled parameters for this experiment were the: applied pressure, sample concentration matrix of simulant material, and pressure surface interactions. This factorial experiment was conducted to determine the effect that the independent parameters of pressure, sampling material, and sample surface have on the dependent variable collection efficiency. The pilot study is shown in the first three columns of Table 6 in the Appendix with three different treatment pressures, five different surface conditions, and one sampling media.

Removable contamination surveys were performed at a single location, multiple times with the same applied pressure. A measure of how much of the sample matrix was removed with each survey was made. The result was then assigned to the specified pressure applied along with the swipe and surface material. The applied pressure was adjusted, and surveys were repeated for each swipe and/or surface material. The resulting data was compiled with 3-dependent variables, analyzed as a factorial experimental design as detailed in Table 6 of Appendix A. This has been done multiple times to understand experimental variation and measure the uncertainty of the experimentation protocol.

To limit the number of changing factors, the area sampled was held constant by controlling the swiping motion of the sampling device to an area of 100 cm². Since the pressure applied to the swipe during sampling was being determined, it was only necessary to measure a pressure applied over a distance. The sampled surface was covered with a dry waste simulant material, supplied by ICP. This simulant material interacts with the IWTU system in the same chemical

manner as the sodium bearing waste that will eventually be introduced. A uniform distribution on the sample surface to be tested was assured by using a roller rail system that compressed a metal sheet into the rail system.



Figure 2 Swipe Sampling Apparatus.

The total amount of simulated contamination material was evenly spread across the testing matrix. The testing matrix was defined by the area beneath the rail system of 14-in length and enclosed by sheet metal barriers. The quantity of areal contamination was determined by distributing a one-tenth gram of simulant per 10 cm² into the matrix sections giving the total amount of contamination to be one gram. Sheet metal cut to the dimensions of the matrix was placed onto the material, pressed, and examined with a level. This procedure ensured a uniform and reproducible distribution within the sample matrix. The simulant contamination material consists of fine particulate. An initial concern was static friction. This environmental factor was controlled by testing in the same relative humidity, room temperature, and other atmospheric conditions.

The cotton swipes were individually weighed to establish a baseline weight for later comparison. The cotton swipes were used based upon the recommendations of 10 CFR 835 Appendix D Table 2-2 as discussed in the literature review section 2.0 Literature Review. The initial sample weight was essential to the gravimetric analysis process as differences in initial weights may alter the calculated removal fractions of the swipes. Therefore, it was imperative to individually measure and record each swipe for accurate evaluation. Weighing each of the samples individually removed uncertainty implied if gravimetric differences in averages were used instead.

The first removal fraction was observed without the use of the apparatus. This was intended to evaluate the variability inherit with manual sampling. Therefore, the manual application of a swipe sampling technique simulated the current protocol for assessing surface contamination. During this simulation, five plausible field surfaces were evaluated with three distinct applied pressures, and one linear swiping pattern. A linear pattern was employed as a representation of the technique to be employed by the IWTU robotic arm. Manual surveying techniques are recommended to be conducted using an "S" shaped swiping pattern according to ANSI N13.4. However, the robotic arm applies pressure in a linear pattern. A linear pattern affords more reproducibility as the same "S" shape is difficult to recreate or imitate mechanically thus, a linear pattern was chosen as the preferrable swiping pattern in this experiment.

The surfaces likely to be encountered by the IWTU sampling operations were investigated. These surfaces were: painted and unpainted concrete, steel, linoleum, and an additional layer of contamination material. The comparison between painted and unpainted concrete is fundamental as the paint is speculated to act as a sealant for a hydrophilic mixture of concrete penetrated by contamination (Sato 2015). Stainless steel is a commonly found material within the nuclear industry with common stainless steel alloys ranging from grade 304 to grade 316 and it is used for the cannisters at IWTU. The polished smooth surface of stainless steel makes it an ideal candidate for decommissioning efforts, as it does not readily incorporate contaminates in to a rough or porous surface. Thus, it was an appropriate model of field conditions. Linoleum was chosen to simulate an observation of contamination in an unexpected area, such as a laboratory floor. The additional layer of material was used to determine if there is a removal factor associated with a buildup of contamination material in one location such as experienced by the leading edge of a swipe.

Three distinct pressures were applied during the manual pilot study of the swipe sampling technique. The first pressure applied was characterized as near zero amount of additional force employed normal to the surface while sampling. The swipe samples taken with near zero applied pressure gravitate towards the surface material by nearly their own weight, not additional force. This minimization of applied pressure was chosen to establish a baseline for other forces to be applied. The second treatment applied was that of moderate pressure. The term moderate pressure is vague, but being the subject of this work, must be explored. Quantification of the phrase is extraneous. A best attempt at field technique was used. This pressure will act as the middle ground for applied pressures and provide a simulation of typical field technique. The applied forces were regulated by weights affixed to the swipe. Specifically, a 1-kg mass was applied to the swipe for moderate pressure of 4.83 Pa for the 0.002 m² swipe medium area. The third distinct pressure applied is the force of overexertion upon the swipe sampling medium, simulated by a 3-kg mass affixed to the swipe of 14.5 Pa. This application of mass will seek to put the maximum amount of force upon the medium without regard to its integrity or efficiency. If swipe integrity degradation is experienced the sample data is lost. The loss of swipe integrity

must therefore be avoided. The overexertion case where swipe integrity is lost acts as the upper bounding limitation to additional force and understanding that value is crucial in optimization of the pressure applied.

The dependent parameter removal fraction (collection efficiency) was evaluated by gravimetric techniques. Weight of each sample was recorded with linearity deviation of ± 0.6 mg. Determining the net weight gain of a sample was accomplished by subtracting the initial weight of the swipe medium from the final weight after the swipe medium has been used to collect a sample. The ratio of contamination found on the swipe per unit area of sample to the total amount of loose contamination present per unit area in the testing matrix is the removal fraction.

The swiping medium is attached to a planchet and placed on the desired surface material within the testing matrix. Cylindrical weights are then placed on top of the planchet to apply the chosen pressure. The weights and swipes are maneuvered by means of the rail system shown in Figure 2 Swipe Sampling Apparatus. This allowed the operator to linearly swipe a contaminated surface within confined testing conditions. Manual application and implementation of the swiping apparatus both analyzed the applied pressures evaluated across each surface material type.

The data from this experiment includes a record of the surface type and amount of pressure applied. The first stage is a manual investigation considering between near zero, moderate, and exaggerated pressures over the unpainted concrete, this was followed by similar tests of painted concrete, steel, and linoleum surface types.

Three trials using a robotic arm were conducted for each surface material type and applied pressure arrangement in the second stage of this investigation. The removal fractions

were documented. Initially, six different pressure treatments were investigated, however, a seventh pressure treatment was added to better establish an optimization curve.

Table 6 of Appendix A provides the recorded data. Histograms of the data sets were plotted showing collection efficiency (the dependent variable) against the (independent variable) applied pressure. These plots were developed for each combination of sample and surface investigated. The estimate of variability in collection efficiency is included in these plots as error bars at plus or minus one sample standard deviation. Uncertainty is assumed to be normally distributed about the mean. The applied pressure in pascals was converted from the mass in grams affixed to pascals of applied pressure by using the mass of the weight and the 2-in diameter swipe area.

4.0 Results and Discussion

The manual application of a linearly swipe sampling technique yielded expected results, with the exception of the unpainted and second layer surfaces experiment. It was anticipated that a linear trend of removal fraction would be correlated with the applied pressure. However, the unpainted and second layer surfaces did not follow a linear trend. A potential explanation for this observation is that the contamination is trapped in porous openings of the surface material by the force applied to the swiping medium, instead of being removed. As the density of the material is low, there is adequate available poor volume for a large fraction of the contamination to be pushed further into the surface rather than adhering to the swiping medium. This explains why a reduction in the removal fraction was not observed in the steel, linoleum, or painted concrete. Deterioration of the swipe integrity was observed during experiments with painted and unpainted concrete surfaces. The frequency of deterioration increased with pressure increases, due to the friction of the swipe on the surface material. It was observed that the exaggerated pressure produced the highest collection efficiency (removal fraction); this is representative of the maximum amount of applied pressure, but it is not necessarily a representation of the optimal removal fraction for each surface type. Although, in these cases it is apparent that the exaggerated applied pressure performed better than applying moderate pressure to the swipe sampling medium. More than moderate pressure may need to be applied to consistently characterize the removable contamination. A graph representing the pilot study results is shown in Figure 3.



Figure 3. Zero, Moderate, and Exaggerated Applied Pressures vs Removal Fraction with Error bars at one sigma.



Figure 4. Removal Fractions with Applied Pressure on a Surface

Figure 4 demonstrates similar characteristics to the preliminary results. Observing the applied pressures over the different surface types on one graph allows one to compare and contrast the removal fractions. The swipes performed on the second layer had the lowest removal fraction performance, while removal fraction performance was observed to be the highest on linoleum. The linoleum has a smooth surface so it is speculated that surface porosity did not trap the contaminants, and thus this smooth surface had low negative affect on the swiping technique, and thus a consistently high removal fraction were observed compared to all other surface material combinations.

Comparisons among the surface material types lead to further insights. The second layer contamination special matieral type was compared with the painted and the unpainted concrete, steel, and linoleum surface types, in the following two figures; Figure 5 and Figure 6.



Figure 5. Second Layer and Unpainted Removal Fractions



Figure 6. Second Layer and Unpainted Removal Overlay

It was observed that for the second layer and unpainted concrete surfaces the removal fraction measured was consistent with those values initially found in the qualitative study. With the application of higher magnitudes of applied pressure, the removal fraction was observed to decrease and then plateau. This characteristic trend is more apparent in the unpainted surface material, as the removal fraction becomes much greater. It was observed to be in the 4,831 Pa to 7,246 Pa range, but then abruptly drops. This was not as apparent in the second layer scenario. It is speculated that this is due to the poor initial removal fraction capabilities. While unpainted or unsealed concrete may not be a common occurrence in contamination areas, caution should be

applied with working on any porous material, it could contain potentially loose contamination fixed within the surface.



Figure 7. Painted, Steel, and Linoleum Removal Fractions



Figure 8. Painted, Steel, and Linoleum Removal Overlay

The next grouping of surface materials were the painted concrete, steel, and linoleum. Each of these surface material types are commonly found in the nuclear industry and thus were appropriate materials for evaluation. The results observed are consistent with the qualitative results for the zero, moderate, and exaggerated pressure test initially performed manually without using the robotic arm. There is a distinct plateau with increased amounts of pressure, indicating a maximum amount of removal fraction possible for a swipe sampling medium. However, the observed trend of a recurring removal fraction with increasing pressure demonstrates a trend of marginal gains, or diminishing returns, with increased effort. It is speculated that this probably reflects a recovery of readily available material, above this, regardless of increased applied pressure other things such as smearing the material into the surface pores, saturation of the sampling medium, or deterioration of the sampling medium compete with contamination recovery by the swipe media.

4.1 Testing Objective Performance

Reproducibility of the swipe sampling technique was evaluated. This was done by evaluating the removal fraction observations for the same applied pressure and surface material combinations. The relative error of population means for these redundant tests were compared to evaluate if this was a reproducible method. The relative error was assessed using a one-sided confidence interval of 95% sample t-test with a cutoff value for determining statistical significance of $\alpha = 0.05$. A p-value of less than or equal to $\alpha = 0.05$ results in rejection of the null hypothesis. A p-value of greater than $\alpha = 0.05$ results in the null hypothesis not being rejected. The p-value is the probability of getting a sample statistic or a more extreme sample statistic in the direction of the alternative hypothesis when the null hypothesis is true. The α level (value) of 0.05 was chosen for the probability of controlling the risk of a Type I error and the t-test was utilized given the number of samples, n, was < 30.

$H_{1,0} =$

The relative error of observations in a group is equal to zero indicating results are reproducible with swipe sampling .

$H_{1,A} =$

The relative error of observations in a group is greater than zero indicating results are not reproducible with swipe sampling.

The relative error of each observation was calculated then the p-values for surface and pressure combinations were calculated using a t-test on the free programming language

developed by the R Foundation for Statistical Computing, R code. Based on the p-values, the null hypothesis is rejected. The alternate hypothesis is supported, and the sampling method is concluded as not reproducible.

Determining optimization of the amount of pressure applied to achieve the greatest representative sample was accomplished by considering the amount of material removed by a swipe sample and its representation of the total known surface contamination.

 $H_{2,0}$ = There is no difference between pressure application for removal fractions.

$H_{2,A} =$ There is a difference between pressure application for removal fractions.

The p-values were calculated using R software, which indicated the removal fractions were distinguishable between the least and most applied pressure. Additionally, comparison of means between applied pressure and removal fraction focused the optimal ranges. Pressure does have an impact on removal for this range of pressures, but we were not able to clearly identify the optimal pressure for each surface given our sample sizes, only that certain ranges of pressure were better than others for different surfaces. Therefore, the null hypothesis is rejected demonstrating the difference in removal fractions from applied pressures. Table 5 considers the applied pressure and the concept of media deterioration. It appears based upon the pairwise t-test and mean interpretation that the optimal applied pressures as seen in Table 5 for each combination of materials considered are between 7,246 and 12,077 pascals.

Applied	1208	2415	4831	7246	8454	9662	12077
Pressure	Pa						
Second	0.66%	0.66%	1.17%	1.17%	0.98%	0.97%	0.88%
Layer							
Unpainted	0.88%	1.46%	2.91%	3.00%	2.14%	1.55%	1.46%
Painted	2.04%	2.83%	2.91%	2.97%	3.20%	3.32%	3.29%
Steel	0.88%	1.46%	1.75%	2.23%	3.10%	3.51%	3.58%
Linoleum	2.43%	2.75%	2.91%	3.33%	3.78%	4.05%	4.06%

Table 5 Removal Fractions for Applied Pressure (Pa) Material Combinations

Based on these measured data, the removal factor as a function of applied pressure is shown in Figure 9.



Figure 9 Removal Fraction Trend Curves for Material Combinations

The last objective was to determine if results from swipe sampling could reliably be used better as a qualitative or a quantitative technique measurement. These data were analyzed by comparing the estimated surface contamination values from the swipe sample to the industry accepted surface contamination values.

 $H_{3,0} =$

There is no difference between the experimental removal factor and the industry accepted removal factor.

 $H_{3,A} =$

There is a difference between the experimental removal factor and the industry accepted removal factor.

All mean removal rates were far less than the 10% industry standard, and the p-value is very small, providing strong evidence for the alternative hypothesis. Additionally, based on these values the industry standards and experimental results differ in greater magnitudes with porous material and less applied pressures. Therefore, the null hypothesis is rejected and there is a difference between the experimentally measured removal fraction and the industry accepted removal factor for estimating total surface contamination.

5.0 Summary and Conclusion

The reproducibility effect of pressure on removal factors, and an evaluation of how swipe sampling may be used as a means of contamination quantification were conducted. The reproducibility of results was not supported with the given applied pressures across each surface material, according to the measured experimental data, highest removal fractions are achieved within the 7,246 to 1,2077 pascal range for painted concrete, linoleum, and steel. This interpretation arises from an observed plateau region. The correlation between applied pressure on a surface material and the removal factor of the swipe sampling medium was found to be statistically significant (p = 0.05) with an optimal weight range of 9,662 to 12,077 pascal for non-porous surface materials and 7,246 to 8,454 pascals for porous materials. The optimal applied pressure, given these caveats are: 7,246, 7,246, 9,662, 12,077, 12,077 pascals for the Second Layer, Unpainted Concrete, Painted Concrete, Stainless Steel, and Linoleum material combinations respectively.

6.0 Future Work

The future work for this project will go into the development of a radio-analytical process for evaluation the swipe samples. Future relevant projects should consider using a combination of radio-analytical and gravimetric methods of assessments.

Investigation into the effects of swipe sampling on porous materials could be a worthwhile endeavor. Even more so if a wetting agent was simultaneously evaluated, to determine its impact on fixed contamination sources.

7.0 References

Cember, H., & Johnson, T. (2009). *Introduction to Health Physics* (Fourth ed.). New Baskerville: The McGraw-Hill Companies, Inc.

Davis, D. M.; Gupton, E. D.; Hart, J. C. *Applied health physics radiation survey instrumentation*. ORNL-332; 1954.

Klein, R. C.; Linins, I; Gershey, E. L. *Detecting removable surface Contamination*. Health Phys. 62186-189; 1992.

Hacker, B.C. *The dragon's tail-radiation safety in the Manhattan Project, 1942-1946.* Berkeley, CA: University of California Press; 1987.

International Organization for Standardization. *Evaluation of surface contamination*part1: beta emitters and alpha emitters. ISO-7503-1; Geneva; 1988

Isamu Sato, Koji Maeda, Mitsuo Suto, Masahiko Osaka, Toshiyuki Usuki & Shin-ichi Koyama (2015) *Penetration behavior of water solution containing radioactive species into dried concrete/mortar and epoxy resin materials*, Journal of Nuclear Science and Technology, 52:4, 580-587, DOI: 10.1080/00223131.2014.963725

Sturchio, G. M.; King, G. R.; Dorman, M. J.; Meechan, P.J. *Adding water to liquid scintillation cocktail for laboratory wipe tests*. Health Phys. 76(Suppl. 1):S32-S36;1999

MARSAME. 2009. Multi-Agency Radiation Survey and Assessment of Materials and Equipment [Manual] (MARSAME), EPA 402-R-06-002.

W. Frame, Paul & Abelquist, Eric. (1999). Use of Smears for Assessing Removable Contamination. Health Physics.76(Supplement 2):S57-S66; 1999

U.S. Department of Energy: 10 CFR Part 835 Occupational radiation protection; final rule. Federal Register 63(213); 1998

U.S. Department of Energy. *Application of DOE 5400.5 requirements for release and control of property containing residual radioactive material.* Memorandum from R.F. Pelletier. OEPA. November 17, 1995.

U.S. Department of Transportation. Radioactive materials. U.S. Code of Federal Regulations 49 CFR 173, Subpart 443. Washington DC: U.S. Government Printing Office; 1992

U.S. Environmental Protection Agency (EPA). *Performance-Based Approach to the Use of Swipe Samples in a Radiological or Nuclear Incident*, Cincinnati, OH, EPA 600/R-11/122, October 2011

U.S. Nuclear Regulatory Commission. *Radiation safety surveys at medical institutions*. Regulatory guide 8.21. Washington DC: U.S.; 1979.

U.S. Nuclear Regulatory Commission. *Radiation safety surveys at medical institutions*. Regulatory guide 8.23. Washington DC: U.S.; 1979.

Appendix A. Removal Fraction Results

		Juito
Surface	Pressure	RF
Unpainted	Zero	0.0146 ± 0.0006
Unpainted	Zero	$0.0088 {\pm}\ 0.0006$
Unpainted	Zero	0.0059 ± 0.0006
Unpainted	Moderate	$0.0291 {\pm}\ 0.0006$
Unpainted	Moderate	$0.0233 {\pm}\ 0.0006$
Unpainted	Moderate	$0.0378 {\pm}\ 0.0006$
Unpainted	Exaggerated	$0.0117 {\pm}\ 0.0006$
Unpainted	Exaggerated	$0.0088 {\pm}\ 0.0006$
Unpainted	Exaggerated	$0.0233 {\pm}\ 0.0006$
Unpainted	1208	$0.0087 {\pm}\ 0.0006$
Unpainted	1208	0.0004 ± 0.0006
Unpainted	1208	$0.0174 {\pm}\ 0.0006$
Unpainted	2415	$0.0145{\pm}\ 0.0006$
Unpainted	2415	0.0062 ± 0.0006
Unpainted	2415	$0.0232{\pm}\ 0.0006$
Unpainted	4831	$0.0289{\pm}\ 0.0006$
Unpainted	4831	$0.0207{\pm}\ 0.0006$
Unpainted	4831	$0.0376{\pm}\ 0.0006$
Unpainted	7246	0.0299 ± 0.0006
Unpainted	7246	$0.0216 {\pm}\ 0.0006$
Unpainted	7246	$0.0386 {\pm}\ 0.0006$
Unpainted	8454	$0.0376{\pm}\ 0.0006$
Unpainted	8454	0.0062 ± 0.0006
Unpainted	8454	0.0203 ± 0.0006
Unpainted	9662	$0.0289 {\pm}\ 0.0006$
Unpainted	9662	$0.0207{\pm}\ 0.0006$
Unpainted	9662	$0.0376{\pm}\ 0.0006$
Unpainted	12077	0.027 ± 0.0006
Unpainted	12077	$0.0265{\pm}\ 0.0006$
Unpainted	12077	$0.0357 {\pm}\ 0.0006$
Painted	Zero	0.0204 ± 0.0006
Painted	Zero	0.0349 ± 0.0006
Painted	Zero	0.0204 ± 0.0006
Painted	Moderate	0.032± 0.0006
Painted	Moderate	$0.0291{\pm}\ 0.0006$
Painted	Moderate	0.0204 ± 0.0006
Painted	Exaggerated	0.0204 ± 0.0006

Table 6 Removal Fraction Results

Painted	Exaggerated	$0.0406{\pm}\ 0.0006$
Painted	Exaggerated	$0.0378 {\pm}\ 0.0006$
Painted	1208	$0.0203{\pm}\ 0.0006$
Painted	1208	0.012 ± 0.0006
Painted	1208	0.0289 ± 0.0006
Painted	2415	$0.0304{\pm}\ 0.0006$
Painted	2415	$0.0187{\pm}\ 0.0006$
Painted	2415	$0.0357{\pm}\ 0.0006$
Painted	4831	$0.0289{\pm}\ 0.0006$
Painted	4831	$0.0207{\pm}\ 0.0006$
Painted	4831	$0.0376{\pm}\ 0.0006$
Painted	7246	0.027 ± 0.0006
Painted	7246	$0.0265{\pm}\ 0.0006$
Painted	7246	$0.0357{\pm}\ 0.0006$
Painted	8454	$0.0318{\pm}\ 0.0006$
Painted	8454	$0.0236{\pm}\ 0.0006$
Painted	8454	$0.0405{\pm}\ 0.0006$
Painted	9662	$0.0203 {\pm}~ 0.0006$
Painted	9662	$0.0329{\pm}\ 0.0006$
Painted	9662	0.0463 ± 0.0006
Painted	12077	$0.0328{\pm}\ 0.0006$
Painted	12077	$0.0245{\pm}\ 0.0006$
Painted	12077	$0.0415{\pm}\ 0.0006$
Steel	Zero	$0.0088{\pm}\ 0.0006$
Steel	Zero	$0.0146{\pm}\ 0.0006$
Steel	Zero	$0.0088{\pm}\ 0.0006$
Steel	Moderate	0.0175 ± 0.0006
Steel	Moderate	$0.032 {\pm}\ 0.0006$
Steel	Moderate	0.0175 ± 0.0006
Steel	Exaggerated	$0.0551 {\pm}\ 0.0006$
Steel	Exaggerated	0.0233 ± 0.0006
Steel	Exaggerated	0.0291 ± 0.0006
Steel	1208	0.0087 ± 0.0006
Steel	1208	0.0004 ± 0.0006
Steel	1208	0.0174 ± 0.0006
Steel	2415	0.0087 ± 0.0006
Steel	2415	0.0004 ± 0.0006
Steel	2415	0.0174 ± 0.0006
Steel	4831	0.0174 ± 0.0006
Steel	4831	0.0091 ± 0.0006
Steel	4831	0.026 ± 0.0006
Steel	7246	0.0222 ± 0.0006
Steel	7246	0.0139 ± 0.0006

Steel	7246	0.0309 ± 0.0006
Steel	8454	$0.0318{\pm}\ 0.0006$
Steel	8454	$0.0207{\pm}\ 0.0006$
Steel	8454	$0.0405{\pm}\ 0.0006$
Steel	9662	$0.0232{\pm}\ 0.0006$
Steel	9662	$0.0358{\pm}\ 0.0006$
Steel	9662	$0.0463 {\pm}\ 0.0006$
Steel	12077	$0.0357{\pm}\ 0.0006$
Steel	12077	0.0274 ± 0.0006
Steel	12077	0.0444 ± 0.0006
Linoleum	Zero	0.0146 ± 0.0006
Linoleum	Zero	$0.0291 {\pm}\ 0.0006$
Linoleum	Zero	$0.0291{\pm}\ 0.0006$
Linoleum	Moderate	$0.0378{\pm}\ 0.0006$
Linoleum	Moderate	0.0262 ± 0.0006
Linoleum	Moderate	$0.0291 {\pm}\ 0.0006$
Linoleum	Exaggerated	$0.058 {\pm}\ 0.0006$
Linoleum	Exaggerated	$0.0435{\pm}\ 0.0006$
Linoleum	Exaggerated	$0.0204{\pm}\ 0.0006$
Linoleum	1208	0.0145 ± 0.0006
Linoleum	1208	0.0207 ± 0.0006
Linoleum	1208	0.0376 ± 0.0006
Linoleum	2415	0.0291 ± 0.0006
Linoleum	2415	0.0243 ± 0.0006
Linoleum	2415	0.0291 ± 0.0006
Linoleum	4831	0.0289 ± 0.0006
Linoleum	4831	0.0207 ± 0.0006
Linoleum	4831	0.0376 ± 0.0006
Linoleum	7246	0.0309 ± 0.0006
Linoleum	7246	0.0293 ± 0.0006
Linoleum	7246	0.0395 ± 0.0006
Linoleum	8454	0.0376 ± 0.0006
Linoleum	8454	0.0293 ± 0.0006
Linoleum	8454	0.0463 ± 0.0006
Linoleum	9662	0.0376 ± 0.0006
Linoleum	9662	0.0492 ± 0.0006
Linoleum	9662	$0.0347 {\pm}\ 0.0006$
Linoleum	12077	0.0405 ± 0.0006
Linoleum	12077	0.0322 ± 0.0006
Linoleum	12077	0.0492 ± 0.0006
Second Layer	Zero	0.0001 ± 0.0006
Second Layer	Zero	0.003 ± 0.0006
Second Layer	Zero	0.003 ± 0.0006

Second Layer	Moderate	0.0059 ± 0.0006
Second Layer	Moderate	0.0175 ± 0.0006
Second Layer	Moderate	$0.0117 {\pm}\ 0.0006$
Second Layer	Exaggerated	0.003 ± 0.0006
Second Layer	Exaggerated	0.0146 ± 0.0006
Second Layer	Exaggerated	0.0088 ± 0.0006
Second Layer	1208	0.0029 ± 0.0006
Second Layer	1208	0.0054 ± 0.0006
Second Layer	1208	0.0116 ± 0.0006
Second Layer	2415	0.0029 ± 0.0006
Second Layer	2415	0.0054 ± 0.0006
Second Layer	2415	0.0116 ± 0.0006
Second Layer	4831	0.0116 ± 0.0006
Second Layer	4831	0.0033 ± 0.0006
Second Layer	4831	0.0203 ± 0.0006
Second Layer	7246	0.0116 ± 0.0006
Second Layer	7246	0.0033 ± 0.0006
Second Layer	7246	0.0203 ± 0.0006
Second Layer	8454	0.0116 ± 0.0006
Second Layer	8454	0.0004 ± 0.0006
Second Layer	8454	0.0174 ± 0.0006
Second Layer	9662	0.0087 ± 0.0006
Second Layer	9662	0.0116 ± 0.0006
Second Layer	9662	$0.0088 {\pm}\ 0.0006$
Second Layer	12077	0.0087 ± 0.0006
Second Layer	12077	0.0004 ± 0.0006
Second Layer	12077	0.0174 ± 0.0006

Appendix B. R Code Plot Examples

R Code Plot Examples

Histogram for Figure 3, Figure 4, Figure 5, and Figure 7

library(ggplot2) library(ggthemes) Values <- data.frame(Surface = $c(\tilde{)}$ Applied = c()Removal = c()myData <- aggregate(Values \$Removal, by = list(cyl = Values \$Surface, gears = Values \$Applied),FUN = function(x) c(mean = mean(x), sd = sd(x),n = length(x)))myData <- do.call(data.frame, myData) myData\$se <- myData\$x.sd / sqrt(myData\$x.n) colnames(myData) <- c("cyl", "gears", "mean", "sd", "n", "se") myData\$names <- c(paste(myData\$cyl, "cyl /", myData\$gears, " gear")) $dodge <- position \ dodge(width = 0.9)$ *limits* <- *aes(ymax* = *myData\$mean* + *myData\$se, ymin* = *myData\$mean* - *myData\$se)* p <- ggplot(data = myData, aes(x = factor(cyl, levels = c("Second Layer", "Unpainted", "Painted", "Steel", "Linoleum"), ordered = TRUE), y = mean, fill = factor(gears, levels = c("1208","2415", "4831", "7246", "8454", "9662","12077"), ordered = TRUE))) $p + theme \ classic() +$ geom bar(stat = "identity", position = position dodge(0.9), colour = "black") + $geom_errorbar(limits, position = position_dodge(0.9), width = 0.25) +$ labs(x = "Surfaces", y = "Removal Fractions") +scale_fill_grey(start = 1, end = 0.4, name = "Applied Pressure (Pa)")

Line Plot for *Figure 6* and *Figure 8*

library(ggplot2) library(ggthemes) Values <- data.frame(Surface = c(), Applied = c(), Removal = c()myData <- aggregate(Values\$Removal, by = list(cyl = Values\$Applied, gears = Values\$Surface), FUN = function(x) c(mean = mean(x), sd = sd(x),n = length(x)))myData <- do.call(data.frame, myData)</pre> myData\$se <- myData\$x.sd / sqrt(myData\$x.n) colnames(myData) <- c("cyl", "gears", "mean", "sd", "n", "se") myData\$names <- c(paste(myData\$cyl, "cyl/", myData\$gears, "gear")) $dodge <- position_dodge(width = 0.9)$ *limits* <- *aes(ymax* = *myData\$mean* + *myData\$se, ymin* = *myData\$mean* - *myData\$se)* p <- ggplot(data = myData, aes(x = factor(cyl, levels = c("1208", "2415", "4831", "7246", "8454", "9662", "12077"), ordered = TRUE), y = mean, group = factor(gears), respectively.shape=factor(gears), linetype=factor(gears))) $p + geom \ errorbar(limits, \ position = position \ dodge(0.9), \ width = 0.25) +$ geom line() + geom point()+ labs(x="Applied Pressure (Pa)", y = "Removal Fraction")+ theme classic()+ theme(text = element text(size = 18, family ="Times"))+ scale shape(name = "Surfaces")+ scale linetype(name = "Surfaces")

Appendix C. R Code Outputs Examples

Example R Code Outputs

1. Reproducibility

> t.test(Data, mu=0, alternative="greater", conf.level=0.95) One Sample t-test data: Data t = 11.154, df = 104, p-value < 2.2e-16 alternative hypothesis: true mean is greater than 0 95 percent confidence interval: 0.2624043 Inf sample estimates: mean of x 0.30827335

2. Pressure Optimization Effectiveness

```
[1] "Pressure" "Removal"
> Fit <- aov(two$Removal ~ two$Pressure)
> summary(Fit)
       Df Sum Sq Mean Sq F value Pr(>F)
two$Pressure 1 0.003054 0.0030538 22.44 6.97e-06 ***
Residuals 103 0.014019 0.0001361
---
Signif. codes: 0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' '1
> pairwise.t.test(two$Removal,two$Pressure)
    Pairwise comparisons using t tests with pooled SD
data: two$Removal and two$Pressure
  250 500 1000 1500 1750 2000
500 1.0000 - - - - -
1000 0.4609 1.0000 - - - -
1500 0.1369 0.7800 1.0000 - - -
1750 0.0862 0.5060 1.0000 1.0000 -
                                  -
2000 0.0091 0.0911 1.0000 1.0000 1.0000 -
2500 0.0089 0.0910 1.0000 1.0000 1.0000 1.0000
```

P value adjustment method: holm

3. Relative to Industry Standards

One Sample t-test

data: Data\$Dif

t = 61.108, *df* = 104, *p*-value < 2.2e-16

alternative hypothesis: true mean is not equal to 0

95 percent confidence interval:

 $0.07392900 \ 0.07888815$

sample estimates:

mean of x

0.07640857