DETERMINATION OF PETROLEUM PIPE SCALE SOLUBILITY IN SIMULATED LUNG FLUID

A Thesis

by

JASON RODERICK CEZEAUX

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

August 2004

Major Subject: Health Physics

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Approved as to style and content by:

Ian S. Hamilton (Chair of Committee)	John W. Poston (Member)
Michael Ryan (Member)	Michael Walker (Member)
William Burchill (Head of Department)	

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ABSTRACT

Determination of Petroleum Pipe Scale Solubility

in Simulated Lung Fluid. (August 2004)

Jason Roderick Cezeaux, B.S., Texas A&M University

Chair of Advisory Committee: Dr. Ian S. Hamilton

Naturally occurring radioactive material (NORM) exists in connate waters and, under the right conditions during oil drilling, can plate out on the interior surfaces of oil and gas industry equipment. Once deposited, this material is commonly referred to as "scale." This thesis is concerned with the presence of ^{226}Ra in scale deposited on the inner surfaces of oil drilling pipes and the internal dose consequences of inhalation of that scale once released.

In the process of normal operation, barium sulfate scale with a radium component adheres to the inside of downhole tubulars in oil fields. When crude flow is diminished below acceptable operational requirements, the pipe is sent to a descaling operation to be cleaned, most likely by a method known as rattling. The rattling process generates dust. This research investigated the chemical composition of that aerosol and measured the solubility of pipe scale from three oilfield formations.

Using standard in-vitro dissolution experimental equipment and methods, pipe scale is introduced into simulated lung fluid over a two-week period. These samples are analyzed using quadrupole inductively coupled plasma mass spectrometry (Q-ICP-MS), known for very low detection limits. Analysis reveals virtually no ^{226}Ra present in the lung fluid exposed to pipe scale. Sample measurements were compared against background measurements using Student's *t* test, which revealed that nearly all the samples were statistically insignificant in comparison to the lung fluid blanks. This

statistical test proves within a 95% confidence interval that there is no ^{226}Ra present in the lung fluid samples. These results indicate that inhaled NORM pipe scale should be classified as Class S and serve to further confirm the extreme insolubility of petroleum pipe scale.

For dose calculations, the S classification means that the lung is the main organ of concern. Radium-226 from petroleum pipe scale does not solubilize in the interstitial lung fluid, and does not, therefore, enter the bloodstream via respiratory pathways. Since there is no removal by dissolution, the 500 day biological half-life implied by the S classification is based solely on the mechanical transport of ^{226}Ra out of the lungs by phagocytosis or the mucociliary escalator.

DEDICATION

To God, Katherine, and my family and friends. Without your love, help, and encouragement, this would not have been possible.

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I wish to express my grateful appreciation to the many people who helped me along the way. To Dr. Ian Hamilton, my advisor and friend; thank you for your ideas and guidance. To Erich Fruchtnicht; together we came to realize that research isn't just a day job. Thank you very much for all the help. Words can't express my appreciation. To Dr. Dennis James; thank you for the use of your characterization lab equipment and your guidance in the use of such. To Dr. Robert Taylor; thank you for your expertise on the exotic topic of ICP-MS. To Robert Berry; thank you for your equipment wrangling. To Dr. Matthew Arno; thank you for your advice and expertise. To Dr. Robert Metzger; thank you for your guidance on NUREG/CR-6419. To the members of my committee, Dr. John Poston, Dr. Mike Ryan, and Dr. Michael Walker; I couldn't have asked for a better committee. Thank you for your prompt responses and willingness to work with me.

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CHAPTER I

INTRODUCTION

PIPE SCALE

Origin of Petroleum Pipe Scale

Over the course of normal oilfield pumping operations, petroleum pipe scale can deposit on the inside of down-hole pipes. Pipe scale consists of concentrated inorganic solids such as barium sulfate, and has been shown to contain naturally occurring radioactive materials (NORM), predominantly compounds of radium.

Naturally occurring radioactive material (NORM) is ever-present in the earth's crust. Uranium and its progeny nuclides leach into subsurface waters and are dissolved along with the barium, calcium, and other dissolved elements. The daughter of concern in this research is ^{226}Ra. ^{226}Ra is an intermediate nuclide in the decay of ^{238}U as shown in Figure 1. ^{226}Ra decays through several nuclides to stable ^{206}Pb, as shown in Figure 2.

Uranium and radium have opposite solubilities in sulfate- and chloride-rich brines. Since chloride remains in solution over the ranges of pH possible in groundwater, chloride leaching can remove radium from a subsurface formation while leaving other nuclides behind (Cowen and Weintritt 1976, Wilson 1992). Under the correct combination of thermodynamic, kinetic, and hydrodynamic conditions, minerals precipitate out of solution and become scale (Hamlat 2003). Because it is chemically analogous (Group II), radium also may become part of the scale by co-precipitation with barium, strontium, and calcium salts (Hamlat 2001). Eventually, micronuclei are formed and begin to agglomerate.

This thesis follows the style and format of Health Physics.

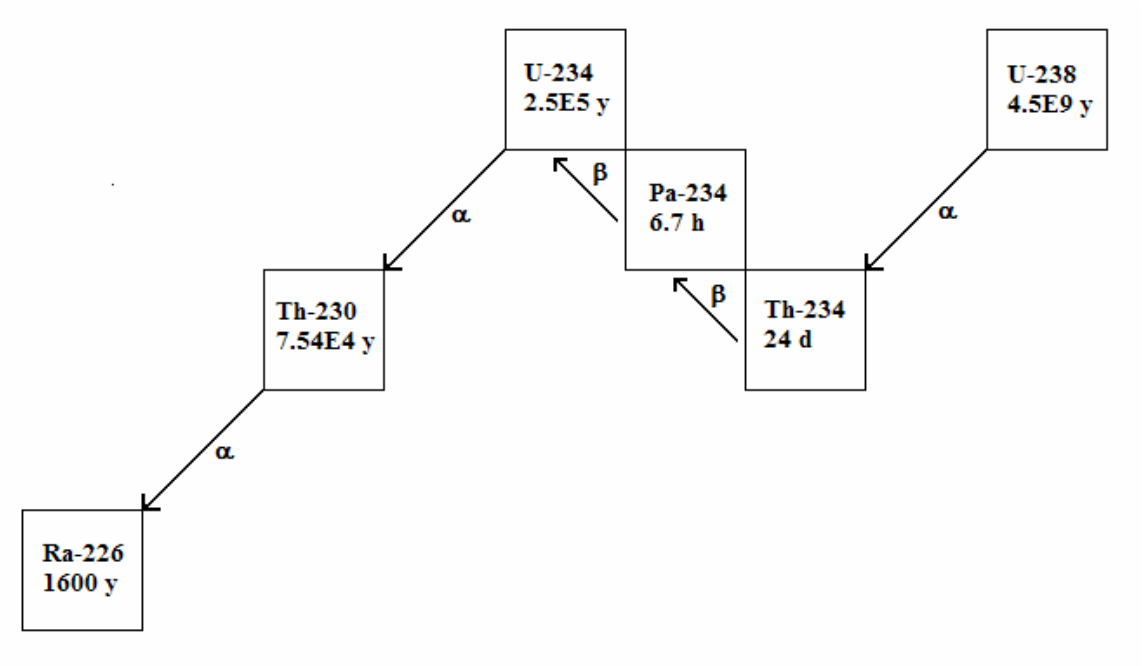

Figure 1. Primary modes of decay for ^{238}U decay to ^{226}Ra, with half-lives of each nuclide listed.

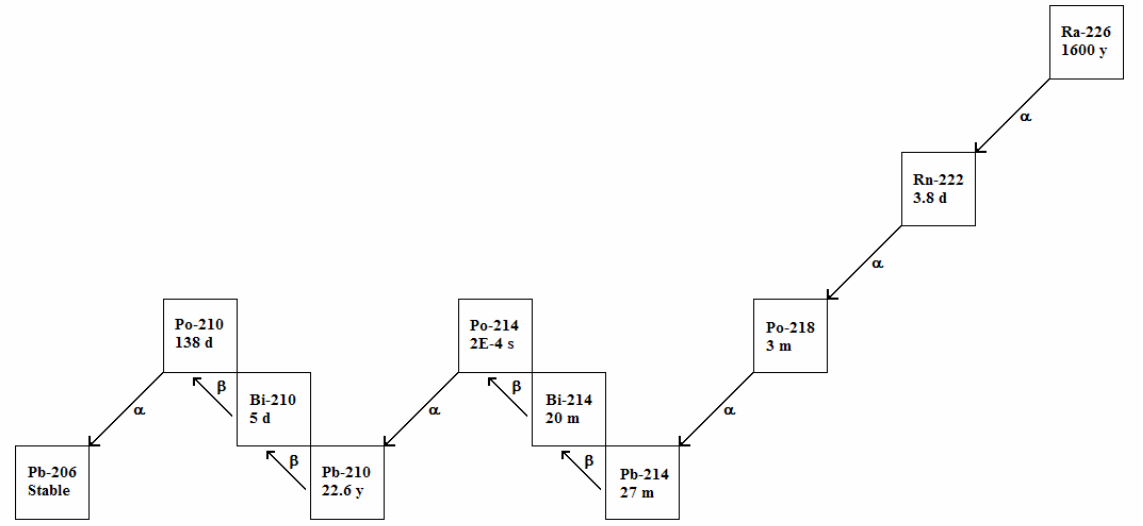

Figure 2. Primary modes of decay for ^{226}Ra decay to ^{206}Pb, with half-lives of each nuclide listed.

Once critical cluster size is attained, scale, in a self-aggravating process, precipitates out and adheres to the pipe interiors (Vetter 1975). Self-aggravation is a process in which, as scale begins to precipitate out, the turbulence caused by the presence of scale causes more scale to precipitate.

When scale buildup occludes oil flow through the pipe, the pipe is removed from the well and sent to be cleaned. Due to the extremely insoluble nature of the scale, chemical cleaning methods cannot be used. In one alternative method of cleaning, the tightly-bound scale is ground out of the pipe with an auger-type bit mounted on an air motor. The bulk of the ground scale is expelled from the opposite end of the pipe, shown in Figure 3, creating a potential airborne radioactive material concern. It is this phenomenon and associated personnel exposure pathways that prompted research in lung fluid solubility as an important part of estimating doses from inhaled pipe scale.

Previous work has been done to determine the solubility of pipe scale in human gastrointestinal fluid (Raabe 1996). Through this work and others, it has been shown that barium sulfate scale is extremely insoluble, even in harsh acidic environments. Based on these results, it is hypothesized that there will be very little dissolution of the scale in simulated lung fluid. A definite classification for the dissolution half-life, the time it takes for half of the inhaled material in the lungs to solubilize, does not exist for pipe scale.

Figure 3. Scale released during dry pipe cleaning experiment.

LUNG DEPOSITION AND MODELING

Deposition in the Lung

Lung deposition and clearance are complex processes that involve many competing chemical and mechanical factors. Clearance from the lung can be achieved via dissolution or mechanical transport. Once inhaled, particles are first deposited in the extracellular airway lining fluid. Particles that are dissolved in this fluid are transferred into the blood. This absorption is the only process easily modeled in-vitro. Particles not dissolved in the interstitial fluid in the first day are phagocytized by macrophages in the airways and alveoli, then held in phagolysosomes (pH=4.5 to 5.5) until either the material is dissolved or the cell dies. Because of the complex cellular transport processes, there is no viable in-vitro test method to determine material movement via this mechanism.

Some percentage of scale particles coming from pipe cleaning processes are respirable (Hamilton, et al. 2004). These particles, once inhaled, are deposited in the lung. The NORM components of the scale are then only biologically available as the barium sulfate crystals are dissolved in the lung.

Lung Models

The use of material solubility has varied as the science has progressed. In 1959, the International Commission on Radiation Protection (ICRP) issued Publication 2, "Report of ICRP Committee II on Permissible Dose for Internal Radiation." This publication did not define the anatomy of the respiratory tract, the kinetics of lung clearance, nor address the dependence of clearance on the solubility of the material. Regardless, the ICRP 2 lung model was used to derive Maximum Permissible Concentrations (MPCs) in air that were used for the next 30 years to calculate dose to a single "critical organ."

In 1966, the Task Group on Lung Dynamics, headed by Owen Moss, developed a new lung model. In 1979, ICRP issued Publication 30, "Limits for Intakes of Radionuclides by Workers" (ICRP 1979). This report was based on the 1966 Task Group on Lung Dynamics and was designed to improve on the ICRP 2 model while retaining its simplicity. The model clearly defined anatomy and kinetics and addressed solubility. Solubility was addressed through the use of Class D, W, and Y. Class D is defined as material that is cleared from the pulmonary region with a biological half-time less than 10 days. Class W is defined as material that is cleared from the pulmonary region with a biological half-time more than 10 days, but less than 100 days. Class Y is defined as material that is cleared from the pulmonary region with a biological half-time more than 100 days. These classifications refer to total clearance (mechanical and absorption to blood). An important change with regard to solubility from ICRP 2 to ICRP 30 was the meaning of "insoluble." In ICRP 2, the term "insoluble" was used to mean that no material reached the blood, and therefore, no material was deposited in any organs or

tissues. In the ICRP 30 model, even some Class Y material is allowed to reach the blood (through compartments a, c, and e).

In 1994, ICRP issued Publication 66, "Human Respiratory Tract Model for Radiological Protection" (ICRP 1994). This lung model was the most complete model to date. The purpose behind the ICRP 66 lung model was to be as realistic as possible. That is, the model was designed to be more realistic than conservative. Anatomy and physiology, including lung afflictions such as smoking and asthma, were addressed in detail for the whole population. In terms of solubility, this model enabled knowledge of the dissolution behavior of specific materials to be used in the calculation of lung dose (ICRP 1994). Mechanical clearance of particles to the lymph nodes and gastrointestinal tract depended on the material. This movement via phagocytosis and the mucociliary escalator was built into the model. Absorption to blood was material-specific and was classified as fast (F), moderate (M), and slow (S). These classifications referred solely to absorption to blood, as particle transport processes and absorption to blood are seen as competitive processes. For this reason, in-vitro lung solubility experiments, which do not attempt to model mechanical clearance, are readily adapted to the F/M/S classification system. One of the major advances of the ICRP 66 lung model was the ability to incorporate time-dependent dissolution data into the model.

Figure 4 shows the anatomical regions of the ICRP 66 respiratory tract model. In this model, ET denotes extrathoracic regions such as the nasal passages, mouth, pharynx, and larynx. BB compartments are the trachea and main bronchi (airway generations 0-8), where deposited material is cleared via ciliary function. The bronchiolar region is indicated by bb; this region consists of the bronchioles and terminal bronchioles (airway generations 9-15). LN denotes lymphatics and lymph nodes that either drain to the extrathoracic region (LN_{ET}) or the thoracic region (LN_{TH}). Finally, AI indicates the alveolar-interstitial region. This region consists of respiratory bronchioles, alveolar

ducts and sacs with their alveoli, and the interstitial connective tissue (airway generations 16 and beyond).

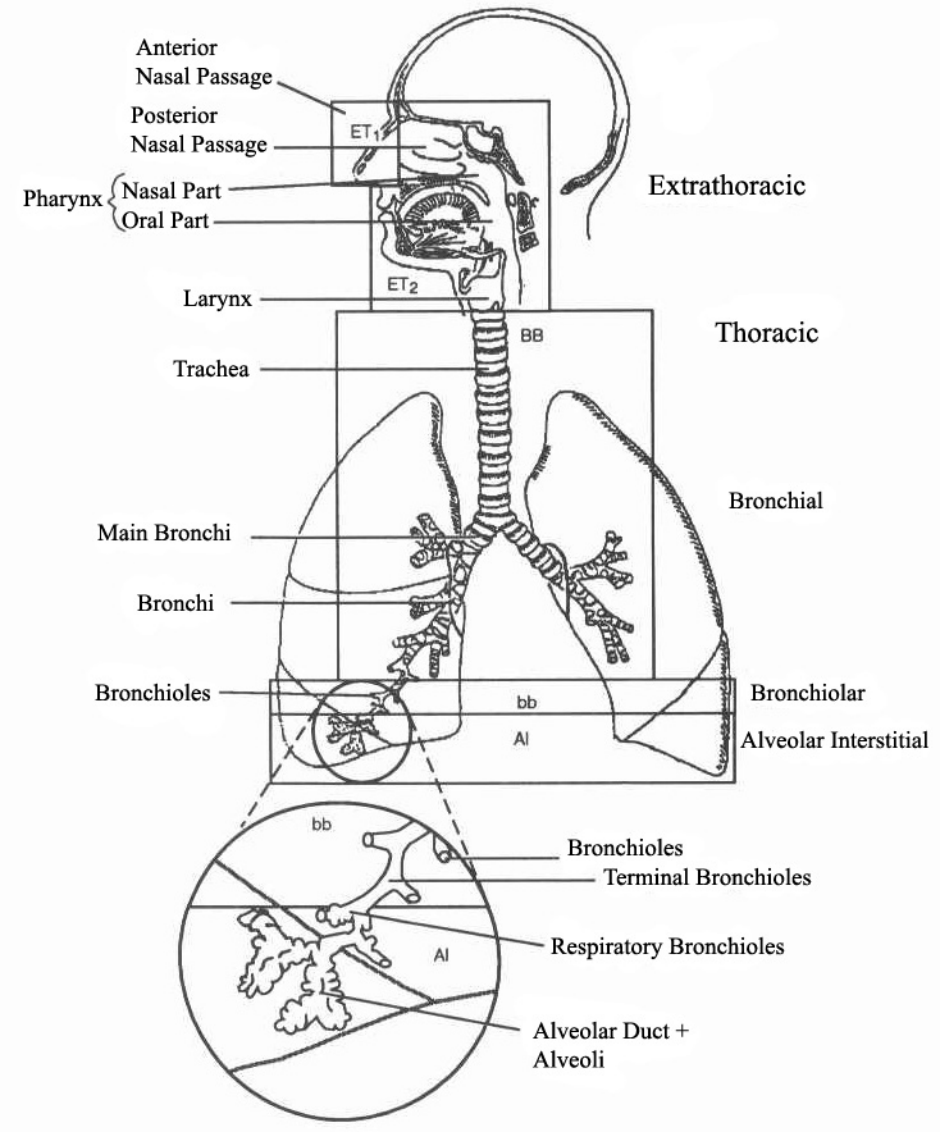

Figure 4. Anatomical regions of the ICRP 66 Respiratory Tract Model (ICRP 1994).

These same regions are utilized in Figure 5, the ICRP 66 overall compartment lung model that shows time-dependent particle transport from each compartment and absorption to blood. This model allows for time-dependent transport rates and absorption. However, most material transport requires no more than a single constant

absorption rate. In this figure, there are three routes that deposited material could take into the blood. These three routes differ by the particle state and stay time. Particles can be absorbed to blood in their initial state or a transformed state, but both states have the same clearance rates (i.e. $(1-f_b)s_p=(1-f_b)s_t$). Bound material will then be absorbed by the blood at its own rate s_b.

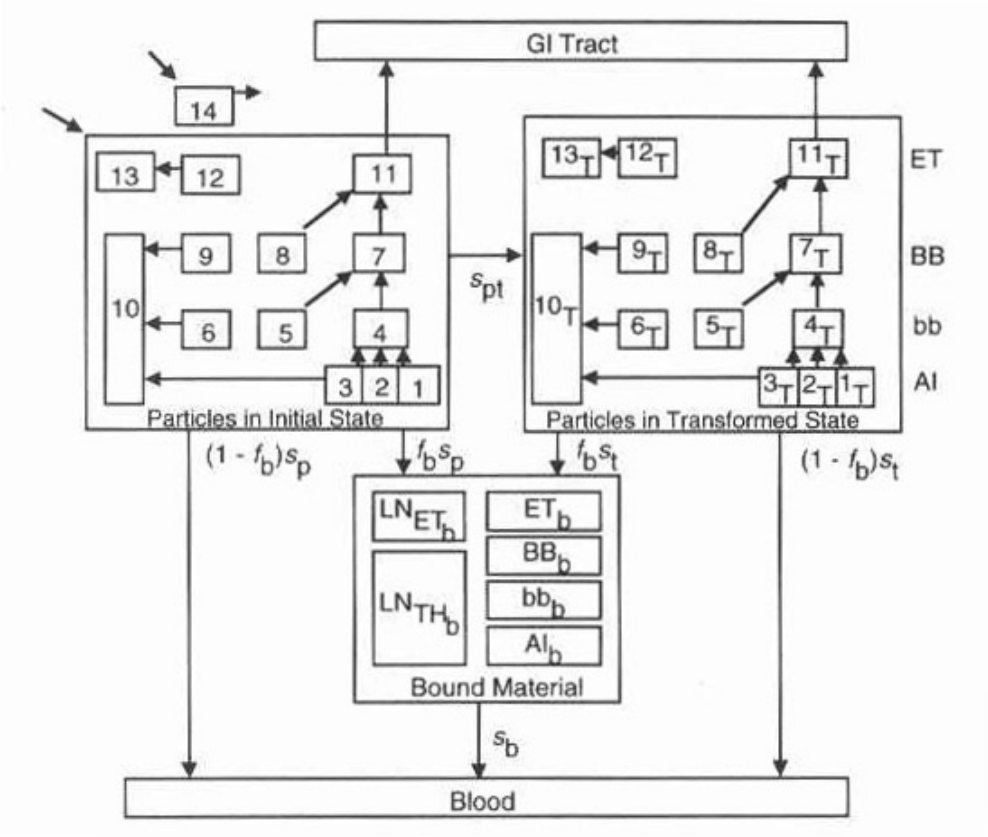

Figure 5. Overall respiratory tract clearance model (ICRP 1994).

The two most common internal radioactive material exposure scenarios are inhalation and ingestion. It is in these two manners that radioactive material may enter the blood stream and distribute throughout the body. In inhalation, material may move from the alveolar-interstitial region into the many pulmonary capillaries there. In ingestion, radioactive material may be absorbed from material that enters the GI tract. Ingestion of radioactive material may occur on its own, but no inhalation intake is solely an

inhalation uptake. Because of mechanical removal from the lung, namely the mucociliary escalator, inhalation exposures are *de-facto* ingestion exposures, as well. The mucociliary escalator moves foreign material to the pharynx to be swallowed, at which point the material enters the GI tract as an ingestion uptake.

Solubility Studies

The many parameters of solubility studies have been optimized through decades of use. This optimization is based on comparison of in-vitro lab results with in-vivo solubility studies when possible. Respiratory tract solubility experiments have been predominantly performed with uranium ore/yellowcake or plutonium oxide dusts. Through a history of trials, certain experimental details have become almost standard.

The first experimental component is the selection of lung fluid stimulant. Owen Moss issued a note in 1979 regarding the best materials to use for serum ultrafiltrate (SUF) (Moss 1979). This work was based on Gamble's solution and has been used ever since. Comparison to the actual constituents of human lung fluid as analyzed by Diem and Lentner shows a very close approximation (Dennis 1982). The material in question is placed into the SUF with filters as a barrier to prevent the sample from dispersing in the solution. This "filter sandwich" is then secured by a Teflon® clamp and submerged in the SUF. The SUF is changed out at a specified time interval (Eidson 1980). The sample may be placed horizontally or vertically in the solution.

Many other experimental variations were addressed by Miglio, et al. in 1977 (Miglio 1977). In this work, they looked at filter position (horizontal vs. vertical), lung fluid movement (static vs. flowing), and fluid amount (100 mL vs. 250 mL). Results of this work showed that these variables did not affect the experimental outcome. The only experimental variables that changed the results were the presence of a polydisperse aerosol and methods in which the aerosol was produced.

Solubility studies should be performed for each specific compound that may be inhaled. One compound of radium, for example, may not dissolve in the same manner as another. This may change their biological half-times and result in over- or under-estimation of dose to the body. This effect has been seen even within the same chemical compound with different isotopes. In his 1987 paper, Ryan found that $^{238}PuO_2$ transferred very differently from $^{239}PuO_2$ (Ryan 1987).

ICP-MS

Inductively coupled plasma mass spectrometry (ICP-MS) has been used as an analysis method for ultratrace detection since the 1980's. This system has become a powerful tool because of its extremely high sensitivity and short measurement times. Additionally, ICP-MS samples require much less chemical preparation than other sampling methods.

Samples go through a rigorous path in ICP-MS. A peristaltic pump channels a sample into the nebulizer, which converts the liquid sample to an aerosol that is suspended in the plasma carrier gas (argon). This aerosol goes through the spray chamber and into the center channel of the plasma torch. The plasma is generated by passing argon through a series of concentric quartz tubes (the plasma torch, shown in Figure 6) that are wrapped at one end by a radiofrequency (RF) coil.

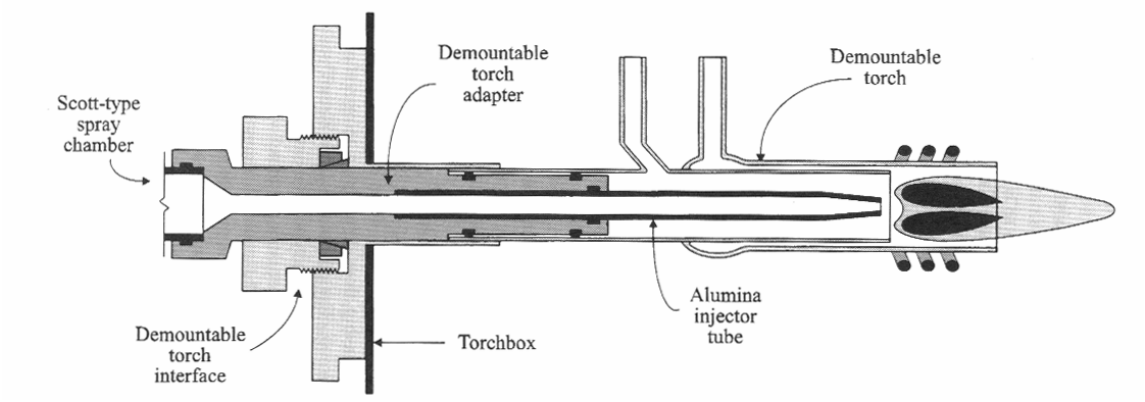

Figure 6. Schematic diagram of a quartz plasma torch. (Montaser 1998)

Energy supplied to the coil by the RF generator couples with the argon to produce the plasma, which operates at a temperature around 6000 °C. Coil layout is shown in Figure 7. As the sample droplets flow through the plasma torch, sufficient energy is added to dry them to a solid, and then sublimate them. The extremely high temperature of the plasma completely breaks apart molecules in the sample. Atoms are ionized in the plasma.

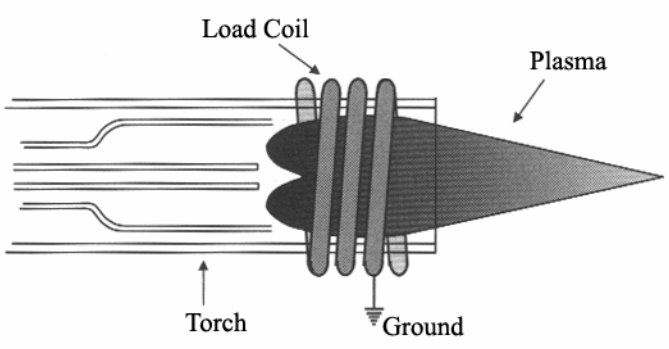

Figure 7. Schematic diagram of the RF coil used to produce an inductively coupled plasma. (Taylor 2001)

After being focused by the ion lens, a charged metallic cylinder, the ions are separated by their mass-to-charge ratio (*m/Z*) in the mass spectrometer. This is accomplished through the use of the quadrupole. The quadrupole mass spectrometer sorts ions by the *m/Z* ratio and allows only one mass to pass through to the detector at any given time. To do this, the quadrupole is set at the correct frequencies to guide ions with the selected *m/Z* between the four poles, while ions with the incorrect *m/Z* are ejected from the quadrupole. A quadrupole analyzer is shown in Figure 8. Despite the fact that the quadupole analyzer only allows one mass through, its rapid scanning speed allows the instrument to see from *m/Z*=1 to *m/Z*=240 in less than 0.1 seconds.

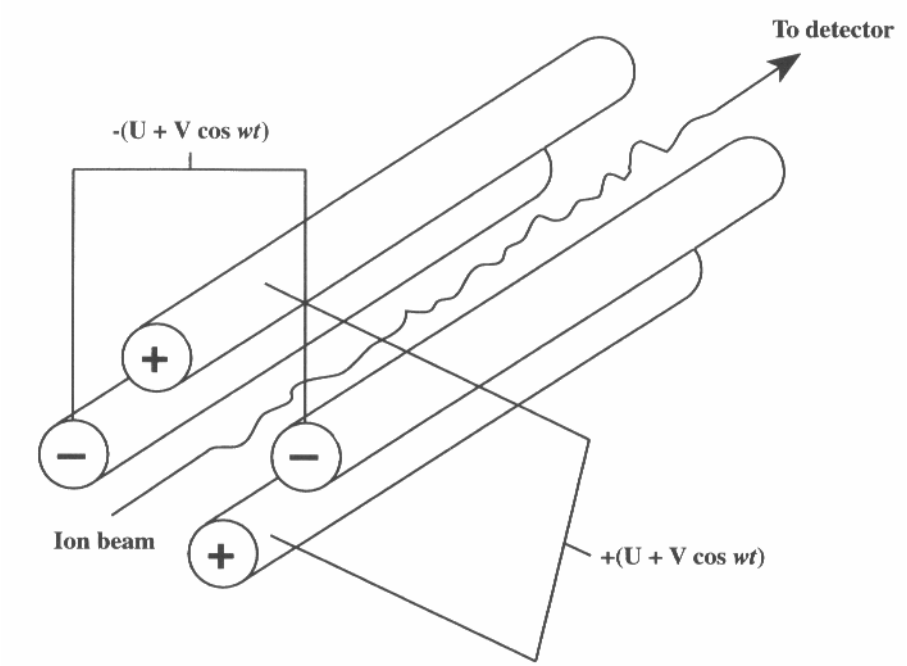

Figure 8. Schematic diagram of a quadrupole mass analyzer. (Montaser 1998)

After the selected ions exit the quadrupole mass analyzer, they are incident on the dynode of the mass detector. As with common photomultiplier tubes, a series of dynodes allows for signal amplification until a measurable pulse is created. The dynode amplification process is shown in Figure 9. Counting these pulses allows calculation of the number of ions that struck the first dynode.

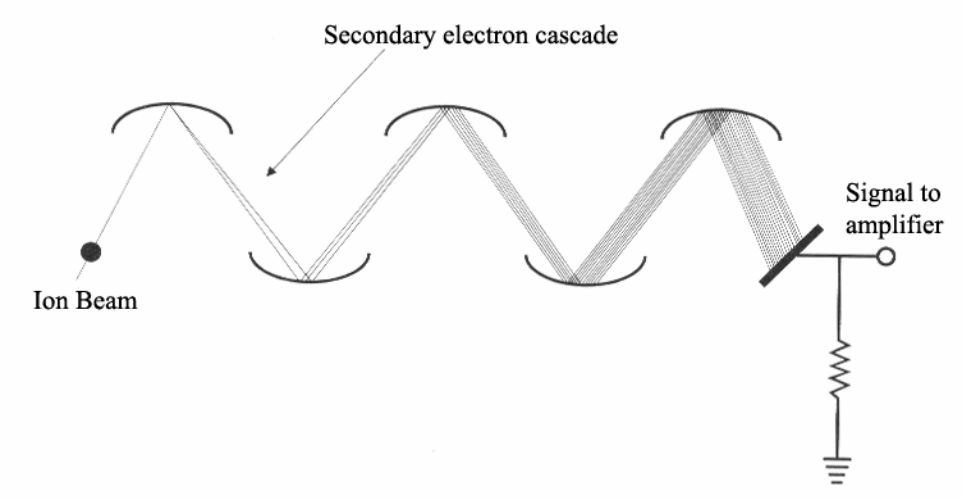

Figure 9. Schematic diagram of a discrete dynode electron multiplier. (Montaser 1998)

In a typical quantitative analysis, liquid standards with known concentrations are analyzed and the results produce a calibration curve. The unknown samples are compared to the calibration curve to determine the unknown concentration.

ICP-MS is very similar to ICP-Optical Emission Spectrometry (ICP-OES) and Graphite Furnace Atomic Absorption (GFAA) in terms of ion production. However, ICP-OES and GFAA systems identify analytes via measurement of emitted light. The ICP-MS system detects and measures the analyte ions themselves. This allows for lower detection limits and the ability to determine individual isotopes of each element.

ICP-MS is not an unproven technique in the detection of radionuclides. When used to measure radionuclide concentrations, ICP-MS has excellent detection limits. In the proceedings of "The Second Symposium on Applications of Inductively Coupled Plasma-Mass Spectrometry to Radionuclide Determinations," held in March of 1998, the American Society for Testing and Materials (ASTM) highlights the use of ICP-MS for detection and quantification of ^{234}U, ^{238}U, ^{99}Tc, ^{237}Np, actinides, and fission products (Morrow 1998). In addition to these papers, numerous others indicate the viability of ICP-MS use in the determination of radionuclides, in which detection limits in the

$fg \cdot mL^{-1}$ range are not unheard of. In the detection of ^{239}Pu, ^{240}Pu, and ^{241}Am, Pointurier et al. obtained results below the fg mL^{-1} range (Pointurier 2004). ICP-MS has also seen use in detection of ^{226}Ra in environmental samples and drinking water (Kim et al. 1999, Hodge 1994). The extremely low detection limits are due to the large masses, and therefore relatively easy identification, associated with many long-lived radionuclides.

CHAPTER II

METHODS AND MATERIALS

DISSOLUTION

Defining the Source Term

To account for the chemical and radiological variance between oilfields, scale samples from three geographically separate oilfields in Texas and Louisiana were analyzed in these procedures. These three formations will be referred to as "Lake Sand" (LS), "Mud Lake" (ML), and "West Delta" (WD).

Understanding particle sizes and what is respirable for inhaled aerosols is important for developing an understanding of an inhalation scenario. Dust released by rattling operations has only a small percentage of respirable particles (Hamilton, et al 2004). It was necessary, therefore, to produce the particles for this research. Using a ball mill, existing scale particles with activity median aerodynamic diameter (AMAD) less than 105 μm were crushed to AMAD < 10 μm. Confirmation of this size reduction was accomplished for all three formations using three methods. A light microscope provided initial evidence of the crushed-particle size. A view of particles under the light microscope is shown in Figure 10. The light microscope images confirmed that the particles were being crushed by the ball mill. More accurate particle size data were then obtained using an environmental scanning electron microscope (ESEM). A view of the scale under the ESEM is shown in Figure 11. The data obtained from the ESEM further confirmed particle diameter, but particle AMAD was needed. To accomplish this, scale samples were analyzed with a Coulter Counter. The results of this analysis showed particle size to be 7.1 ± 0.4 μm AMAD.

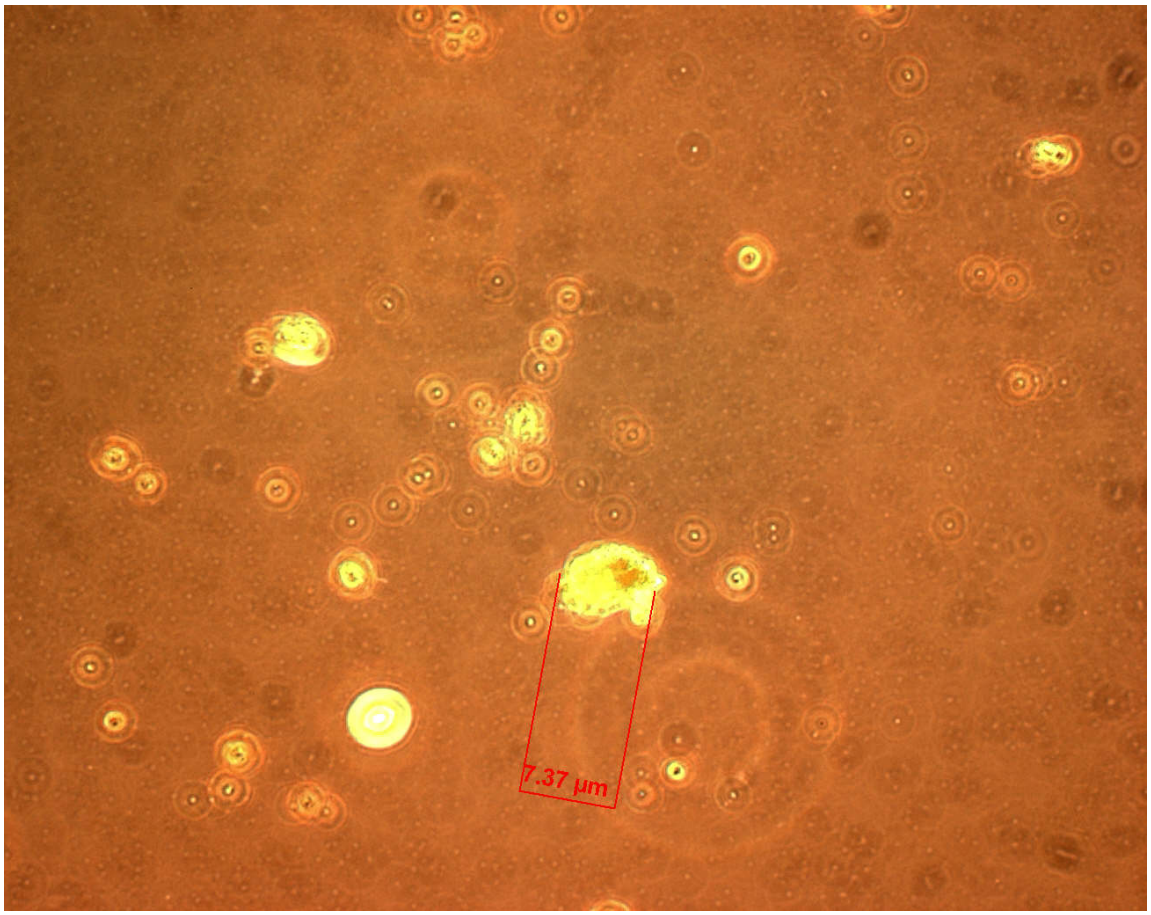

Figure 10. Pulverized scale particles under a light microscope.

Once confirmed to be respirable, the scale particles were analyzed via x-ray fluorescence (XRF) to determine elemental composition. This analysis confirmed large amounts of barium, along with sulfur, calcium, and strontium, or $(BaCaSr)SO_4$. A typical example of results obtained through the use of XRF is shown in Figure 12. The iron peaks in this spectrum are indicative of rust from inside the pipe. Radiochemistry results obtained from the Radioanalytical Branch of the Radiation Surveillance Division at Brooks Air Force Base showed ^{226}Ra concentrations of 910 ± 10 pCi g^{-1}, 1.8 ± 0.02 nCi g^{-1}, and 1.6 ± 0.01 nCi g^{-1} in scale from Lake Sand, Mud Lake, and West Delta, respectively. These results are included in Appendix C.

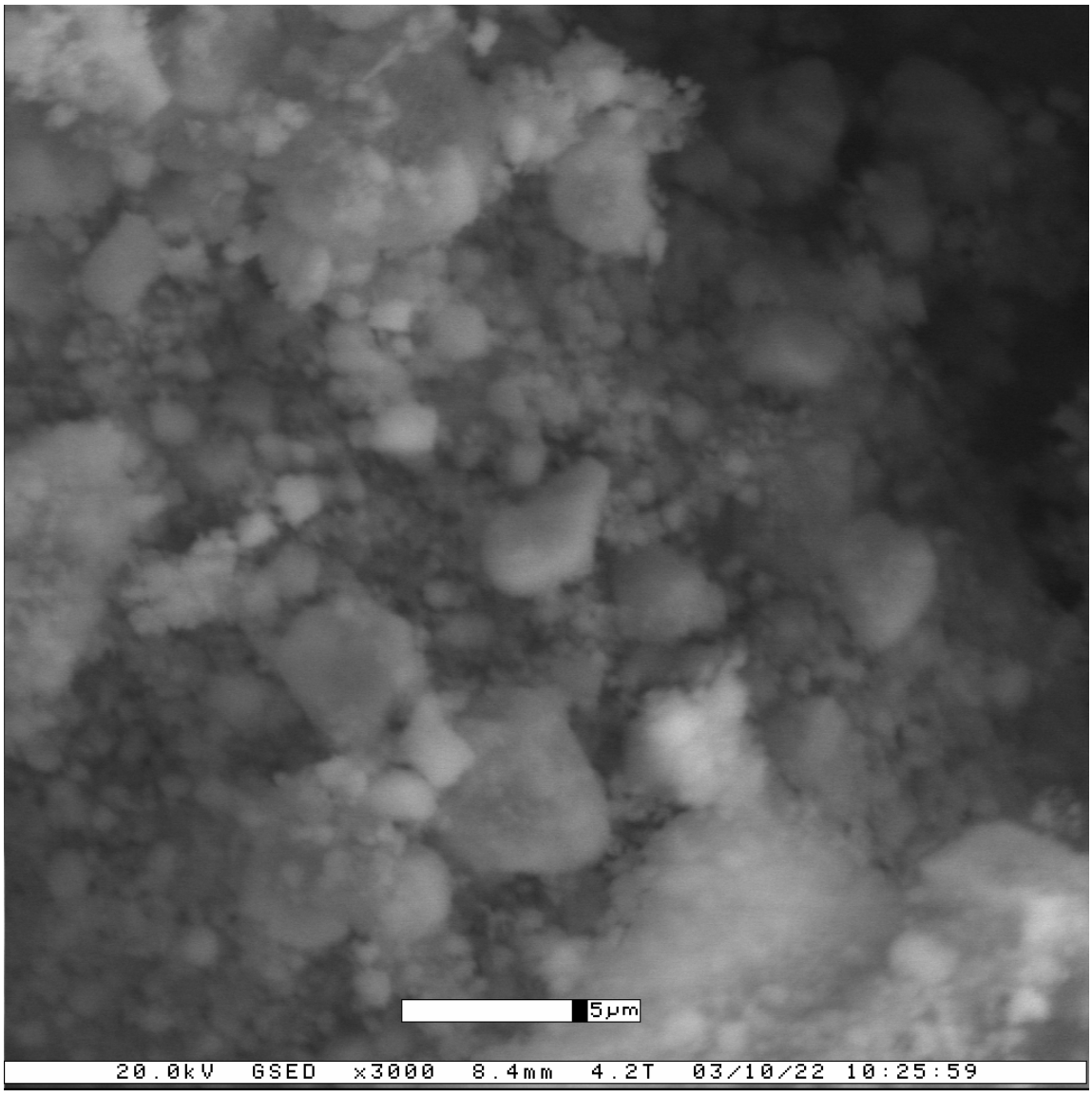

Figure 11. Scanning electron micrograph of pulverized petroleum pipe scale particles used in this solubility study.

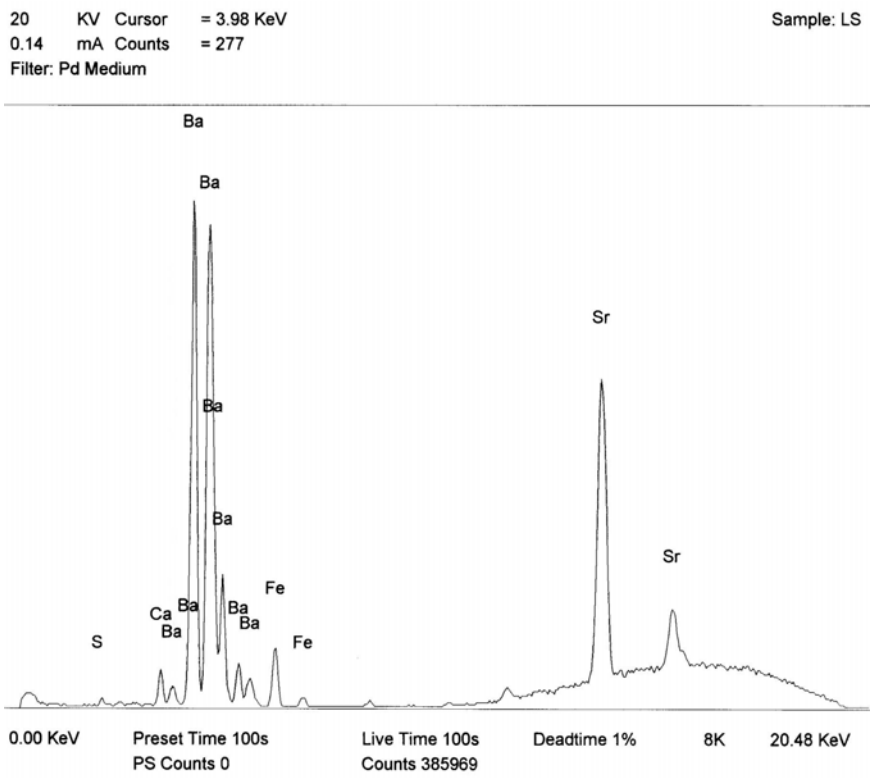

Figure 12. An example of a spectrum obtained from the XRF process.

In ICRP 30, a "standard" particle size, that on which dose calculations are based, is 1 μm AMAD. In ICRP 66, this standard particle size is 5 μm AMAD. Attempts were made to pulverize the scale down to this range, but this was not possible with the available ball mill. This fact speaks to the tremendous energy and work that are required to break this material down into smaller, respirable particles. Another interesting observation is that the physical diameter closely approximates the AMAD of the respirable particles.

In-Vitro Model

Once the source term was defined, the dissolution in the human lung was modeled by using a lab simulant for serum ultrafiltrate (SUF). Serum ultrafiltrate (lung fluid) serves as part of an air-blood barrier in the lung. For an inhaled particle to get into the blood, it must dissolve through this barrier. It is exactly for this reason that the solubility of pipe scale in lung fluid is of concern.

Knowing how long it takes to get from the lungs to the blood is important for dose calculations. Once the material solubilizes in the lung fluid, it enters the bloodstream and can deposit in other organs. In this case, if the ^{226}Ra entered the bloodstream, it would relocate into the bones, causing the bones (and eventually the red bone marrow) to receive dose. If, however, the material does not enter the bloodstream, the lung remains the primary organ of concern.

Lung fluid dissolution trials have been done for many years. Most work in the past has been centered on uranium yellowcake or plutonium solubility. In these previous works, a standard simulant for SUF was developed. The simulated SUF was based on the work of Gamble (1967) and for this experiment, the ingredients and mixing procedure were referenced from NUREG/CR-6419 (Metzger 1996). Molar concentrations of chemicals in the SUF are listed in Table 1.

Table 1. Chemical composition of simulated SUF.

Chemical	Molar Concentration
NaCl	0.116 M
NH_4Cl	0.010 M
$NaHCO_3$	0.027 M
Glycine	0.005 M
L-cysteine	0.001 M
Na Citrate	0.0002 M
$CaCl_2$	0.0002 M
H_2SO_4	0.0005 M
NaH_2PO_4	0.0012 M
DTPA	0.0002 M
ABAC	50 ppm

Two of these chemicals, DTPA and ABAC, are not present in actual serum. Diethylenetriaminepentaacetic acid (DTPA) is a chelating agent used to decrease the amount of dissolved actinide ions sticking to the walls of the dissolution container. Alkylbenzyldimethyl ammonium chloride (ABAC) is used as an antibacterial agent. All salts used in the SUF were reagent grade and the water was distilled and deionized. In addition to matching the chemical make-up of lung fluid, temperature and pH were controlled to match body conditions of 37 °C and pH=7.3.

To create the interface between the scale and the SUF, standard dissolution equipment was used (Kanapilly et al. 1973, Miglio et al. 1977). Equipment setup and sampling followed NUREG/CR-6419 (Metzger 1996), a published procedure concerning solubility testing for actinides on air filters. Scale was placed on top of a 37 mm, 0.4 μm pore size Gelman GN-4 filter, and covered with another clean filter to create a "filter sandwich." The total radium activity on the filters ranged from 16.8 Bq to 32.8 Bq. This sandwich was placed in the filter holder assembly shown in Figure 13. This holder was oriented vertically in 200 mL of static SUF with a constant gas flow of 5% CO_2 to maintain pH. Both the beaker and the filter holder were made of non-corrosive plastic (Teflon®) and the beaker was covered to prevent evaporation losses. This setup is shown in Figure 14. This solution was changed out every hour for the first day, every day for the first week, and every week for the remainder of the month.

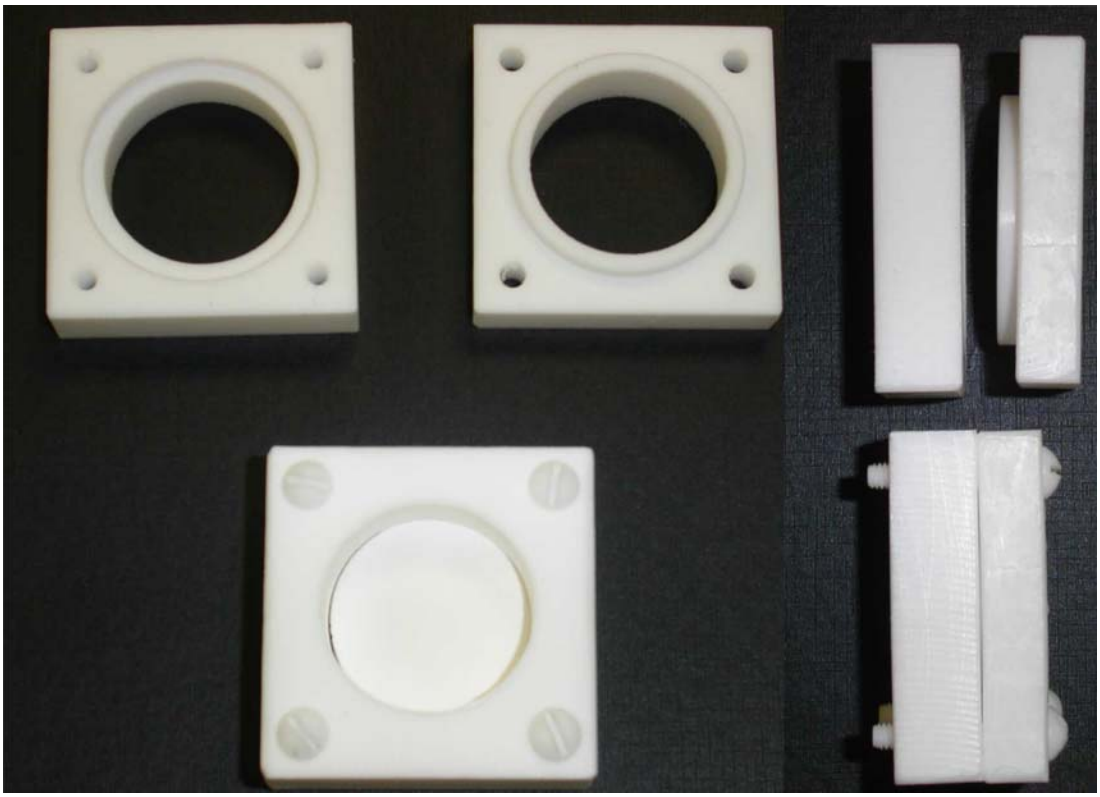

Figure 13. Teflon® filter holder (unassembled and assembled).

One problem that was encountered was that the filters did not hold up to a full month of submersion in the lung fluid. After two weeks, each filter ruptured, cutting the experiment short by two weeks. It was hypothesized that the ruptures were due to mechanical stress caused by the filter holders. Attempts to change the holder configuration did not solve this problem. Caution was taken to end the dissolution trial as soon as signs of rupture were seen so as to prevent loose scale from getting into the lung fluid samples.

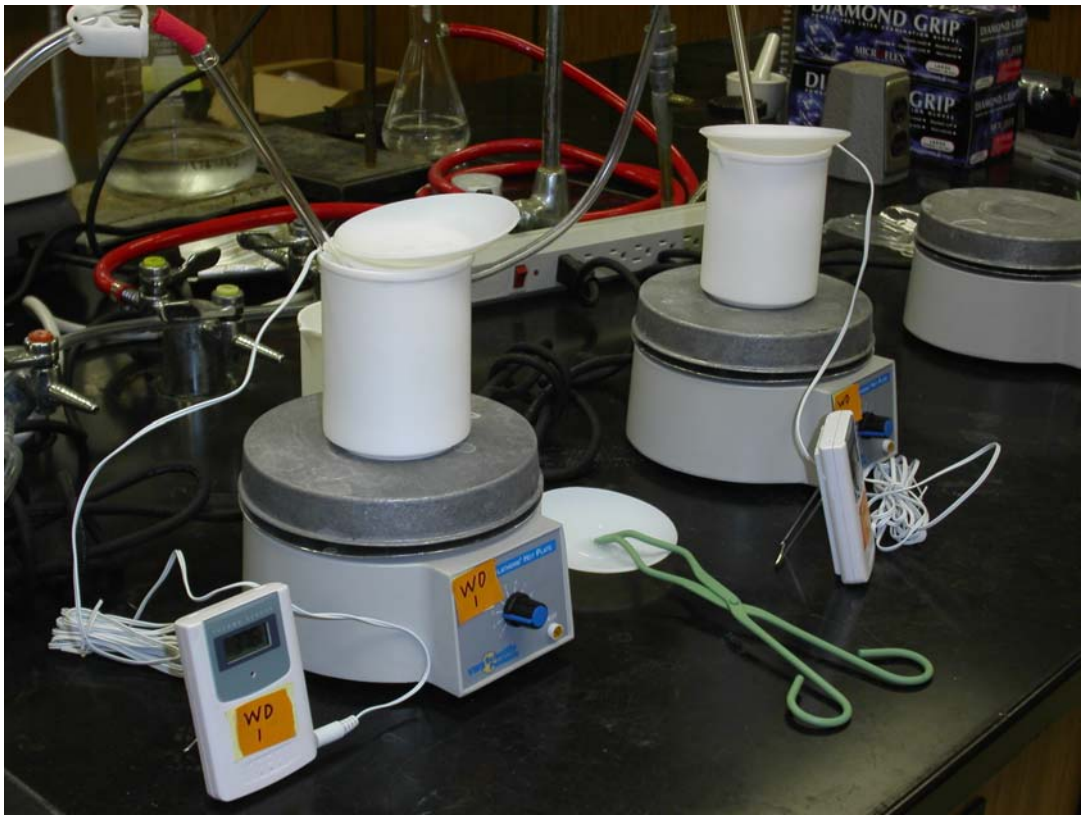

Figure 14. Dissolution experiment setup.

ANALYSIS

ICP-MS Setup

Once the dissolution trials were completed, the lung fluid samples were analyzed for ^{226}Ra by quadrupole inductively coupled mass spectrometry (Q-ICP-MS). ICP-MS measurements were performed with an "ELAN DRC II" (PerkinElmer SCIEX) equipped with a quadrupole mass spectrometer. The solution was pumped by a peristaltic pump (internal three channel pump) and the nebulizer was of the concentric quartz Meinhold-type. The aerosol produced was directed through a quartz cyclonic spray chamber into a quartz plasma torch. Additional argon was supplied to the torch as coolant and as extra support for the plasma. The plasma was maintained at 1400 W. A concise listing of machine parameters is in Table 2. Blank and standard solutions prepared with 1% HNO_3

were used to determine the sensitivity and stability of the system under normal conditions. The ICP-MS was calibrated with uranium standard solutions.

Table 2. ICP-MS Parameters

RF Forward Power	1400 W
Argon gas flow	
Plasma	15 L min^{-1}
Auxiliary	1.2 L min^{-1}
Nebulizer	0.95 L min^{-1}
Sampling Cone	Ni, 1.0 mm aperture
Skimmer Cone	Ni, 0.75 mm aperture
Nebulizer Type	Concentric Quartz Meinhard-type
Solution Uptake	1.3 mL min^{-1}
Dwell Time	50 ms
Number of Scans	6
Channels	1
Scanning mass	226.025 *m/Z*

The sensitivity levels at *m/Z*=226 and 238 were assumed to be the same for two reasons. First, the ionization potentials are 5.277 keV for radium and 6.1 keV for uranium, so both are low enough to assume that ionization differences are not significant in the argon plasma. Second, both masses are relatively close together and in the region of the mass spectrum that shows little mass bias. Using this calibration, it was determined that the detection limit for the analysis was 226 fCi.

Background

Since the ICP-MS system had such high sensitivity, ultrapure reagents, standards, and blanks were used: ultrapure HNO_3 (J.T. Baker), barium stock plasma standard solutions 10 mg L^{-1} (Specpure, Alfa Aesar), multi-element stock standard solution containing ^{238}U (TEXASAM-STD-2A, Inorganic Ventures, Inc.), and ultrapure water with a resistivity of 18.2 MΩ·cm (Milli-Q, Millipore). Each sample was run with an internal standard solution containing ^{209}Bi and ^{103}Rh (6020ISS, Inorganic Ventures, Inc.) to correct for any variation in the ICP-MS signal caused by environmental changes and environmental effects. Internal standards also serve to confirm normal operation from sample to sample. Measurements of the internal standards should stay relatively constant within a sample group. Internal standard measurement confirmations are given in Appendix B.

Sampling

The lung fluid samples contained 10,000 ppm of salts. To prevent suppression of the signal by the abundant salts in solution, each sample was diluted to 10% of the original strength with 1% HNO_3 (Olivares 1986). Rinsing with blank solutions between sample measurements was employed to suppress any memory effect and to avoid any cross-contamination by the sample probe.

To gain a more complete data set, each sample was analyzed in pulse mode by ICP-MS six times. These repetitions are called "replicates." Additionally, each scale formation was run in duplicate, resulting in two dissolution data sets per formation.

ICP-MS vs. PERALS

The original scope of this project was to use Photon-Electron Rejecting Alpha Liquid Scintillation (PERALS). PERALS is an advanced chemical procedure that involves sample concentration via co-precipitating ^{226}Ra, metathesizing the sulfate to carbonate, dissolving the carbonate, and extracting the radium from this solution. The sample is

then counted on a very low background detector that uses pulse shape discrimination to reject counts from incident γ and β radiation.

Being so chemistry-intensive, PERALS samples take on the order of six hours to prepare. The sample then counts in the detector for an hour. ICP-MS samples required five minutes of preparation, and six replicate measurements were made in four minutes. The speed of throughput through ICP-MS and the lack of dependence on advanced chemistry procedures while maintaining a low detection limit made it the preferable analysis procedure.

CHAPTER III

RESULTS

The software associated with the ICP-MS system reports the mean of six replicates from each sample along with the relative standard deviation (RSD). The raw data obtained from the ICP measurements have been included in Appendix A. These spreadsheets also include the statistical analyses.

RADIUM

Examination of Data

Upon examination of the ICP-MS analysis results, the ^{226}Ra concentrations in the samples were comparable to those in the lung fluid samples that had not been exposed to scale. The initial goal was to obtain a dissolution curve based on the amount of dissolved ^{226}Ra in the SUF sample. Since it appeared that there were no results above the detection limit of 226 fCi, statistical analysis was used to confirm whether or not the samples exposed to scale were statistically significant in comparison to the blank SUF samples.

Statistical Significance

Student's *t* test allows for the determination of statistical significance. This statistical test is a comparison of analytical results order to confirm whether both samples contain the same amount of the measured analyte. By comparing the post-treatment sample to the mean background, a confidence interval can be ascertained. The background mean and standard deviation were calculated using the weighted mean and standard deviation formulas shown below (Cember 1996). First, the weighting factor is determined by:

$$w_i = \frac{1}{\sigma_i^2}.$$

The weighted mean is then defined as:

$$M_w = \frac{\sum w_i M_i}{\sum w_i}.$$

The standard deviation associated with this mean is then given by:

$$\sigma_{M_w} = \sqrt{\frac{1}{w_1 + w_2 + \ldots + w_n}}.$$

For the blank samples, the weighted mean was 2.96 ± 0.288 cps.

The sample results were then compared to this weighted mean and standard distribution. Student's t test is performed by first calculating the relative error for a given confidence interval, τ_{calc} (Martin 2000):

$$\tau_{calc} = \frac{|r_1 - r_2|}{\sqrt{\sigma_1^2 + \sigma_2^2}} \text{ where,}$$

r_1 and r_2 are count rates and σ_1 and σ_2 are the standard deviations of r_1 and r_2, respectively. This value is then compared with a true probability value τ_{table}. This τ_{table} value has a probability *p* associated with it, which corresponds to a given confidence interval.

One way to conduct Student's *t* test is to try to confirm a null hypothesis. In this method, one selects a desired confidence interval and determines the associated τ_{table}. The null hypothesis states that the sample, in this case, contains no ^{226}Ra with some given degree of certainty. Once the confidence level has been selected, the associated τ_{table} is obtained from a table. If the null hypothesis is to hold (i.e. there is no ^{226}Ra), τ_{calc} must be less than or equal to τ_{table}, which is dependent on the selected confidence interval. In the analysis of the SUF samples, the selected confidence interval was 95%. This *p* corresponds to a τ_{table} of 1.96 (Martin 2000).

Application of Statistics

Upon application of these statistical tests, the sample results were overwhelmingly insignificant when compared to the background counts. These results are shown in Appendix A. The fact that the samples were not significant, instead confirms that no ^{226}Ra had dissolved into the lung fluid to a certainty of 95%.

Out of the 206 samples analyzed, 198 of them were considered statistically insignificant with a 95% level of confidence. The other 8 samples, or 3.88% of the total samples, that were deemed statistically significant by Student's *t* test, fall within the 5% error associated with the 95% confidence interval. If all the samples had proven to be statistically insignificant using the 95% confidence interval, a positive bias would have been suspected.

BARIUM

Barium Dissolution

In addition to ^{226}Ra, barium was included as an analyte in the ICP-MS analysis. The results showed a dissolution profile much like what would be expected for a relatively insoluble material: a rapid initial dissolution followed by a shallow, asymptotic curve. A barium dissolution curve is shown in Figure 15. It may seem incongruent that while chemically analogous, barium and radium solubilize differently. The fact that barium goes into solution while ^{226}Ra does not is attributable to the fact that $BaSO_4$ actually dissolves more readily than $RaSO_4$. The solubility of $BaSO_4$ in water is 0.00031 g/100 g H_2O while $RaSO_4$ is insoluble in water (Lide 2003). Additionally, there is much more barium than radium in the scale due to these differing solubilities affecting precipitation in the scale deposition process. This preferential deposition also creates a non-uniform matrix in which there is much more barium than radium.

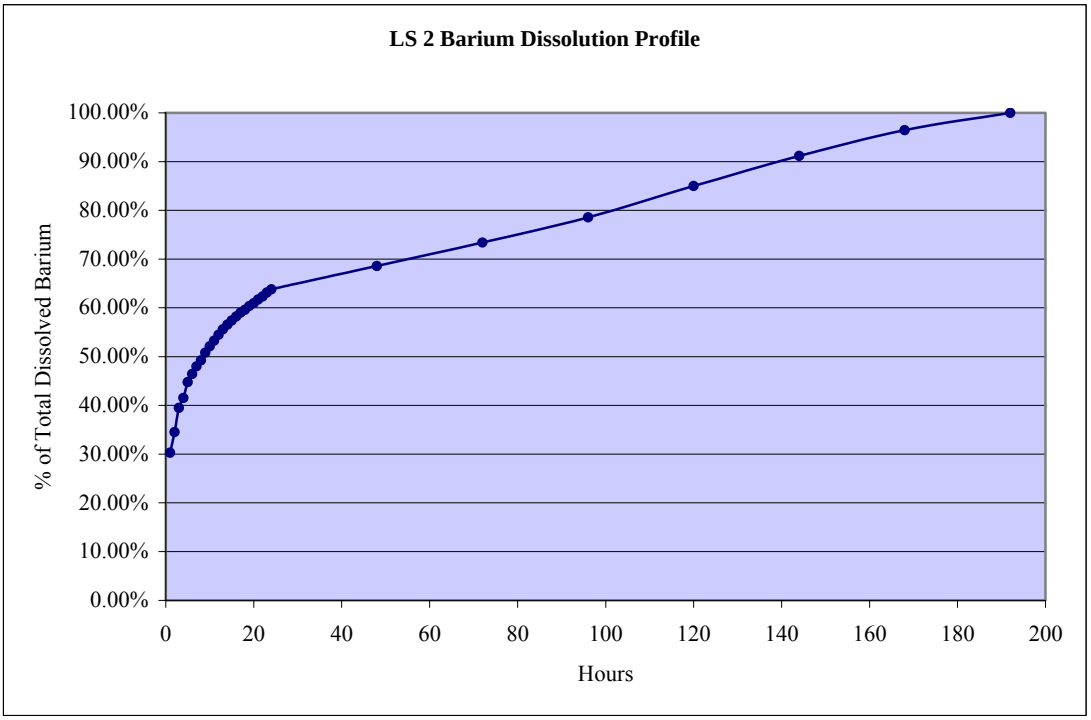

Figure 15. Curve showing the dissolution profile of barium released from scale.

CHAPTER IV
DISCUSSION

The result that there was no ^{226}Ra present in the lung fluid confirms the current assumption of solubility class S. Once scale is inhaled, the ^{226}Ra is not solubilized in the interstitial fluid of the lung, and therefore is not transferred to the blood by absorption in the alveolar-interstitial region of the lung. Therefore, the only removal of inhaled 226Ra from the lung is via mechanical clearance to the GI tract or lymph nodes. This is because no inhalation scenario is isolated to inhalation. Due to the mechanical transport of material out of the lung by the mucociliary escalator and subsequent swallowing, inhalation exposures become ingestions. In the case of ^{226}Ra, the material that moves to the GI tract via swallowing is still very insoluble, as previous work has shown (Raabe 1996). Because of this, the material that does make it to the GI tract is excreted relatively quickly, resulting in very little dose to those organs. Slow removal from the lungs and rapid removal from the GI tract via excretion result in the identification of the lungs as the main organ of concern when assessing internal dose due to ^{226}Ra inhaled in petroleum pipe scale.

The physical processes associated with foreign particle removal from the lungs, the mucociliary escalator and phagocytosis, are the principle methods of ^{226}Ra removal since it is not soluble. As previously mentioned, it is very challenging to develop in-vitro models of these processes. For this reason, ICRP 66 gives mechanical removal rates based on phagocytotic activity. This activity is represented as a component in the dissolution half-life of the material. In the case of class S material, this half-life is assumed to be 500 days.

CHAPTER V

SUMMARY

Pipe scale is a routine occurrence in the drilling industries, particularly in the petroleum industry. The scale that is plated onto the drilling tubulars and other equipment typically contains ^{226}Ra plated out as a sulfate compound. When the tubulars are cleaned, these radioactive materials are released as dust and pose a potential inhalation hazard.

Using simulated serum ultrafiltrate based on Gamble's solution, the dissolution of respirable-sized pipe scale particles in interstitial lung fluid was measured. The lung fluid samples were then analyzed for ^{226}Ra using inductively coupled plasma mass spectrometry.

The results from the ICP-MS analysis showed no dissolution of the ^{226}Ra into the SUF. Statistical analyses (Student's *t* test) revealed that the samples that had been exposed to pipe scale were not statistically different from the lung fluid blanks. Since there was no dissolution, ^{226}Ra in petroleum pipe scale should be classified as solubility class S.

The results obtained by the ICP-MS analysis lead to more analysis opportunities. For future studies, it may be beneficial to analyze exposed lung fluid for thorium, bismuth, lead, and polonium just to see if any of these ^{238}U progeny dissolve in the lung fluid.

It is clear from this study that the NORM radionuclides found in the pipe scale particles are tenaciously held by the particle matrix and that the particles are extremely insoluble in the interstitial fluid of the lung. This leads to the conclusion that ^{226}Ra inhaled during pipe-descaling operations can be classified as class S material, that having exceptionally slow absorption into the blood from the lungs. Therefore, the lung is the main organ of concern when calculating doses attributed to the inhalation of ^{226}Ra in petroleum pipe scale.

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APPENDIX A

RAW DATA AND CALCULATION SHEETS

Lake Sand Run 1

Initial Radium [pCi] = 455.09
Initial Radium [Bq] = 16.838

In Sample:	Hours	Mean Intensity [cps] ^{103}Rh	^{138}Ba	^{209}Bi	^{238}U	^{226}Ra	Intensity % RSD	RSD	Student's *t* test σ	τ_{calc}	τ_{calc} < 1.96?
LS 01	1	177196	1846857	107907	2079	6	43.680	0.437	2.621	1.155	Y
LS 02	2	181477	1506095	107327	1521	6	36.416	0.364	2.185	1.381	Y
LS 03	3	180655	660096	108612	1266	7	31.539	0.315	2.208	1.817	Y
LS 04	4	169056	400284	97642	1275	6	35.319	0.353	2.119	1.424	Y
LS 05	5	177421	323077	105457	1225	6	56.171	0.562	3.370	0.900	Y
LS 06	6	182512	197237	107789	1198	6	12.394	0.124	0.744	3.818	N
LS 07	7	184141	215838	108243	1170	5	26.342	0.263	1.317	1.516	Y
LS 08	8	179114	187887	105917	1179	5	30.407	0.304	1.520	1.321	Y
LS 09	9	183590	137137	109691	1159	4	23.333	0.233	0.933	1.069	Y
LS 10	10	180603	98459	105748	1277	6	40.428	0.404	2.426	1.246	Y
LS 11	11	183068	79803	107312	1289	5	42.400	0.424	2.120	0.956	Y
LS 12	12	176751	126843	100599	1227	4	32.826	0.328	1.313	0.777	Y
LS 13	13	182180	139063	103937	1175	5	26.442	0.264	1.322	1.511	Y
LS 14	14	189763	108397	109551	1137	5	28.029	0.280	1.401	1.429	Y
LS 15	15	192800	100322	111651	1132	4	50.821	0.508	2.033	0.509	Y
LS 16	16	181665	427286	102831	1151	6	9.658	0.097	0.579	4.705	N
LS 17	17	185015	97208	105790	1153	5	30.897	0.309	1.545	1.301	Y
LS 18	18	170168	152747	93722	1155	5	40.505	0.405	2.025	0.999	Y
LS 19	19	192131	90529	110861	1112	6	27.276	0.273	1.637	1.832	Y
LS 20	20	195799	103509	112763	1125	5	31.408	0.314	1.570	1.281	Y
LS 21	21	183111	94447	105871	1122	5	42.359	0.424	2.118	0.957	Y
LS 22	22	187523	101093	109745	1110	6	48.376	0.484	2.903	1.044	Y
LS 23	23	177461	103778	101423	1154	5	32.746	0.327	1.637	1.230	Y
LS 24	24	186114	77103	108387	1098	3	27.143	0.271	0.814	0.052	Y
LS 25	48	182232	611696	105871	1122	4	33.767	0.338	1.351	0.756	Y
LS 26	72	182593	768072	105594	1134	4	28.782	0.288	1.151	0.880	Y
LS 27	96	176827	1139632	100457	1164	4	43.412	0.434	1.736	0.593	Y
LS 28	120	186211	684305	107803	1097	5	41.078	0.411	2.054	0.986	Y
LS 29	144	184902	1004443	105951	1105	5	45.569	0.456	2.278	0.890	Y
LS 30	168	177452	1234089	99714	1107	4	25.330	0.253	1.013	0.992	Y
LS 31	192	184104	789120	104828	1101	5	26.619	0.266	1.331	1.501	Y

Lake Sand Run 2

Initial Radium [pCi] = 455.64
Initial Radium [Bq] = 16.859

In Sample:	Hours	Mean Intensity [cps] ^{103}Rh	^{138}Ba	^{209}Bi	^{238}U	^{226}Ra	Intensity % RSD	RSD	Student's *t* test σ	τ_{calc}	$\tau_{calc} < 1.96$?
LS2 01	1	151088	3366853	87394	5023	3	56.218	0.562	1.687	0.026	Y
LS2 02	2	156606	492787	92034	1447	4	21.082	0.211	0.843	1.172	Y
LS2 03	3	152303	555249	87457	1346	4	37.033	0.370	1.481	0.692	Y
LS2 04	4	162028	246790	94848	1298	5	19.869	0.199	0.993	1.977	N
LS2 05	5	152774	356093	85848	1338	4	31.702	0.317	1.268	0.803	Y
LS2 06	6	168369	208561	97736	1259	5	18.592	0.186	0.930	2.101	N
LS2 07	7	167223	193457	97079	1251	4	39.278	0.393	1.571	0.654	Y
LS2 08	8	165536	152266	95810	1243	5	28.183	0.282	1.409	1.422	Y
LS2 09	9	168294	198460	97553	1214	5	50.067	0.501	2.503	0.811	Y
LS2 10	10	167466	160358	97152	1232	5	30.696	0.307	1.535	1.309	Y
LS2 11	11	163604	140960	95049	1227	5	27.590	0.276	1.380	1.451	Y
LS2 12	12	162580	147271	93968	1257	4	39.722	0.397	1.589	0.647	Y
LS2 13	13	168291	136955	97185	1263	4	47.140	0.471	1.886	0.548	Y
LS2 14	14	165846	117500	95204	1263	5	19.064	0.191	0.953	2.053	N
LS2 15	15	166991	105234	96571	1228	5	57.104	0.571	2.855	0.712	Y
LS2 16	16	167000	103025	97098	1238	3	48.736	0.487	1.462	0.030	Y
LS2 17	17	165953	96038	96245	1771	5	25.206	0.252	1.260	1.582	Y
LS2 18	18	166993	75552	97318	1774	5	48.705	0.487	2.435	0.834	Y
LS2 19	19	164452	97934	95684	2402	4	35.890	0.359	1.436	0.713	Y
LS2 20	20	168255	74471	98818	1921	4	47.410	0.474	1.896	0.545	Y
LS2 21	21	163027	95030	96485	3201	5	29.212	0.292	1.461	1.373	Y
LS2 22	22	163926	79367	98238	1995	5	51.608	0.516	2.580	0.787	Y
LS2 23	23	163313	100299	97388	3808	4	26.924	0.269	1.077	0.937	Y
LS2 24	24	162901	84958	97072	3823	5	40.375	0.404	2.019	1.003	Y
LS2 25	48	165975	604990	99745	1827	4	37.915	0.379	1.517	0.677	Y
LS2 26	72	170988	637591	103607	3873	6	22.934	0.229	1.376	2.166	N
LS2 27	96	165298	647038	98201	3169	3	24.987	0.250	0.750	0.055	Y
LS2 28	120	160987	766265	94120	4161	3	36.553	0.366	1.097	0.039	Y
LS2 29	144	164012	771005	97624	3066	4	49.375	0.494	1.975	0.523	Y
LS2 30	168	162618	650849	97028	3264	5	42.681	0.427	2.134	0.949	Y
LS2 31	192	156899	420772	92366	3522	6	29.342	0.293	1.761	1.707	Y
LS2 32	360	145237	1310255	94767	3004	4	63.775	0.638	2.551	0.407	Y

Mud Lake Run 1

Initial Radium [pCi] = 884.65
Initial Radium [Bq] = 32.732

		Mean Intensity [cps]							Student's *t* test		
In Sample:	Hours	^{103}Rh	^{138}Ba	^{209}Bi	^{238}U	^{226}Ra	Intensity % RSD	RSD	σ	τ_{calc}	$\tau_{calc} < 1.96$?
ML 01	1	153718	4413011	90699	1221	4	22.305	0.223	0.892	1.114	Y
ML 02	2	150440	1521798	87959	1119	4	38.205	0.382	1.528	0.672	Y
ML 03	3	160505	1147047	95877	1098	4	51.307	0.513	2.052	0.504	Y
ML 04	4	160886	306941	95132	1121	5	42.583	0.426	2.129	0.952	Y
ML 05	5	166492	443436	100688	1082	6	32.625	0.326	1.958	1.539	Y
ML 06	6	159646	440137	94735	1099	4	43.128	0.431	1.725	0.597	Y
ML 07	7	163129	247183	98838	1086	5	53.495	0.535	2.675	0.760	Y
ML 08	8	159259	212910	94956	1089	4	43.918	0.439	1.757	0.587	Y
ML 09	9	156702	155198	92601	1095	4	48.804	0.488	1.952	0.529	Y
ML 10	10	157364	180808	93116	1137	6	20.553	0.206	1.233	2.404	N
ML 11	11	161637	140179	98090	1111	5	33.415	0.334	1.671	1.206	Y
ML 12	12	157598	205469	95393	1114	4	38.996	0.390	1.560	0.659	Y
ML 13	13	158404	277845	96094	1094	4	46.123	0.461	1.845	0.559	Y
ML 14	14	159426	190077	97783	1092	4	22.906	0.229	0.916	1.088	Y
ML 15	15	160958	178955	98983	1085	5	40.989	0.410	2.049	0.988	Y
ML 16	16	144610	401195	83628	1151	4	39.344	0.393	1.574	0.653	Y
ML 17	17	158836	239866	96336	1094	4	40.704	0.407	1.628	0.632	Y
ML 18	18	163587	311093	99168	1078	5	24.648	0.246	1.232	1.615	Y
ML 19	19	158748	148904	95846	1133	3	37.022	0.370	1.111	0.039	Y
ML 20	20	163140	117077	99625	1072	5	50.170	0.502	2.509	0.810	Y
ML 21	21	155599	109251	95706	1087	5	30.271	0.303	1.514	1.327	Y
ML 22	22	157908	107734	97994	1075	4	38.920	0.389	1.557	0.660	Y
ML 23	23	158207	89342	98819	1077	3	30.726	0.307	0.922	0.046	Y
ML 24	24	155760	100249	95923	1079	5	33.806	0.338	1.690	1.192	Y
ML 25	48	161339	85201	100787	1082	3	67.811	0.678	2.034	0.022	Y
ML 26	72	152549	4254419	92695	1182	4	21.564	0.216	0.863	1.149	Y
ML 27	96	157050	5159437	96109	1062	4	50.121	0.501	2.005	0.516	Y
ML 28	120	159845	3759611	98028	1064	4	52.945	0.529	2.118	0.489	Y
ML 29	144	166970	2814200	103957	1041	3	50.420	0.504	1.513	0.029	Y
ML 30	168	157658	8582462	95596	1040	5	43.149	0.431	2.157	0.939	Y
ML 31	192	162176	6926590	98605	1066	7	22.583	0.226	1.581	2.517	N

Mud Lake Run 2

Initial Radium [pCi] = 885.53
Initial Radium [Bq] = 32.765

		Mean Intensity [cps]							Student's *t* test		
In Sample:	Hours	^{103}Rh	^{138}Ba	^{209}Bi	^{238}U	^{226}Ra	Intensity % RSD	RSD	σ	τ_{calc}	τ_{calc} < 1.96?
ML2 01	1	151161	1915116	99225	2546	4	55.202	0.552	2.208	0.469	Y
ML2 02	2	149659	629246	96943	1538	4	31.153	0.312	1.246	0.817	Y
ML2 03	3	148485	331765	96158	1343	5	45.586	0.456	2.279	0.890	Y
ML2 04	4	151852	230873	99452	1286	5	38.561	0.386	1.928	1.049	Y
ML2 05	5	148911	199204	96584	1277	3	65.185	0.652	1.956	0.023	Y
ML2 06	6	155203	199262	101358	1244	4	48.469	0.485	1.939	0.533	Y
ML2 07	7	152373	185224	98162	1232	5	37.354	0.374	1.868	1.082	Y
ML2 08	8	151872	165317	96929	1284	5	55.116	0.551	2.756	0.738	Y
ML2 09	9	147986	190103	92892	1262	3	27.217	0.272	0.817	0.051	Y
ML2 10	10	157345	124798	100797	1220	4	30.192	0.302	1.208	0.841	Y
ML2 11	11	153866	164323	98035	1218	3	24.926	0.249	0.748	0.056	Y
ML2 12	12	154472	185988	99538	1243	4	44.163	0.442	1.767	0.584	Y
ML2 13	13	154381	186951	100183	1275	3	40.154	0.402	1.205	0.036	Y
ML2 14	14	155029	113515	98292	1224	3	60.074	0.601	1.802	0.024	Y
ML2 15	15	159982	133363	101598	1233	3	65.393	0.654	1.962	0.022	Y
ML2 16	16	158985	126441	100618	1257	4	51.651	0.517	2.066	0.501	Y
ML2 17	17	158975	124343	100651	1844	3	53.033	0.530	1.591	0.028	Y
ML2 18	18	160308	113657	101762	1756	3	46.579	0.466	1.397	0.031	Y
ML2 19	19	157657	130256	98801	2587	4	41.648	0.416	1.666	0.618	Y
ML2 20	20	157653	104507	98940	2222	4	40.881	0.409	1.635	0.629	Y
ML2 21	21	160839	91154	104078	2897	3	44.695	0.447	1.341	0.032	Y
ML2 22	22	158284	98234	100132	2277	3	41.723	0.417	1.252	0.035	Y
ML2 23	23	153947	103837	97241	4760	3	25.972	0.260	0.779	0.054	Y
ML2 24	24	152254	106711	94951	5008	3	51.952	0.520	1.559	0.028	Y
ML2 25	48	159194	610907	100877	1957	4	36.452	0.365	1.458	0.703	Y
ML2 26	72	163012	760901	105297	2071	5	37.698	0.377	1.885	1.072	Y
ML2 27	96	154516	842191	96909	3286	5	28.822	0.288	1.441	1.391	Y
ML2 28	120	155550	975220	97147	2882	4	44.569	0.446	1.783	0.578	Y
ML2 29	144	154725	999014	95938	3350	5	32.325	0.323	1.616	1.245	Y
ML2 30	168	157581	983837	98000	2891	4	38.930	0.389	1.557	0.660	Y
ML2 31	192	162384	361278	102424	3031	5	22.389	0.224	1.119	1.769	Y

West Delta Run 1

Initial Radium [pCi] = 802.72
Initial Radium [Bq] = 29.701

		Mean Intensity [cps]							Student's *t* test		
In Sample:	Hours	^{103}Rh	^{138}Ba	^{209}Bi	^{238}U	^{226}Ra	Intensity % RSD	RSD	σ	τ_{calc}	$\tau_{calc} < 1.96$?
WD 01	1	190411	264464	136269	1071	5	51.378	0.514	2.569	0.791	Y
WD 02	2	159684	871086	102548	1233	3	52.887	0.529	1.587	0.028	Y
WD 03	3	156917	630049	99588	1196	3	19.693	0.197	0.591	0.068	Y
WD 04	4	161112	415551	103111	1165	4	31.746	0.317	1.270	0.802	Y
WD 05	5	156658	287224	98973	1158	4	36.398	0.364	1.456	0.704	Y
WD 06	6	157947	307611	99814	1238	4	48.048	0.480	1.922	0.537	Y
WD 07	7	155023	169684	97775	1177	4	38.273	0.383	1.531	0.671	Y
WD 08	8	159589	156805	101211	1162	4	68.059	0.681	2.722	0.382	Y
WD 09	9	160828	108379	102198	1135	4	53.639	0.536	2.146	0.483	Y
WD 10	10	154437	147485	96154	1147	3	44.402	0.444	1.332	0.033	Y
WD 11	11	153064	116125	97112	1155	4	27.438	0.274	1.098	0.921	Y
WD 12	12	158053	106070	100244	1134	4	33.576	0.336	1.343	0.760	Y
WD 13	13	155375	104500	97894	1129	3	39.884	0.399	1.197	0.036	Y
WD 14	14	153928	116181	96570	1137	3	20.917	0.209	0.628	0.065	Y
WD 15	15	153780	135004	96291	1251	4	73.364	0.734	2.935	0.354	Y
WD 16	16	152551	156207	95201	1095	4	55.195	0.552	2.208	0.469	Y
WD 17	17	153620	158162	96688	1110	4	44.621	0.446	1.785	0.578	Y
WD 18	18	153671	127106	96483	1101	3	67.980	0.680	2.039	0.022	Y
WD 19	19	152871	121954	95624	1114	3	31.529	0.315	0.946	0.045	Y
WD 20	20	154428	82795	96933	1123	4	21.206	0.212	0.848	1.166	Y
WD 21	21	152665	162448	98178	1100	3	50.723	0.507	1.522	0.029	Y
WD 22	22	149617	196196	94540	1129	3	44.036	0.440	1.321	0.033	Y
WD 23	23	148336	216720	92086	1091	4	28.098	0.281	1.124	0.900	Y
WD 24	24	147971	226907	92801	1158	4	26.169	0.262	1.047	0.962	Y
WD 25	48	150003	731545	92960	1083	3	54.509	0.545	1.635	0.027	Y
WD 26	72	154319	804438	97726	1086	3	33.678	0.337	1.010	0.042	Y
WD 27	96	155259	877866	97868	1083	5	22.513	0.225	1.126	1.760	Y
WD 28	120	157343	1001932	98793	1108	4	27.017	0.270	1.081	0.934	Y
WD 29	144	143011	1566398	86658	1142	3	27.824	0.278	0.835	0.050	Y
WD 30	168	155395	1186075	96740	1102	3	54.037	0.540	1.621	0.027	Y
WD 31	192	153136	1235764	96681	1102	4	17.705	0.177	0.708	1.366	Y

West Delta Run 2

Initial Radium [pCi] = 801.28
Initial Radium [Bq] = 29.647

In Sample:	Hours	Mean Intensity [cps] ^{103}Rh	^{138}Ba	^{209}Bi	^{238}U	^{226}Ra	Intensity % RSD	RSD	Student's *t* test σ	τ_{calc}	τ_{calc} < 1.96?
WD2 01	1	152629	1736973	98922	1299	4	20.411	0.204	0.816	1.207	Y
WD2 02	2	157526	1410904	103401	1163	4	24.748	0.247	0.990	1.013	Y
WD2 03	3	155424	972505	100918	1148	3	37.417	0.374	1.123	0.038	Y
WD2 04	4	155063	1131878	99173	1165	4	55.516	0.555	2.221	0.466	Y
WD2 05	5	156281	832146	101433	1151	3	40.550	0.406	1.217	0.036	Y
WD2 06	6	150724	713015	95399	1277	3	80.183	0.802	2.405	0.018	Y
WD2 07	7	153721	359446	98634	1195	3	49.215	0.492	1.476	0.030	Y
WD2 08	8	154540	231767	98989	1157	3	59.719	0.597	1.792	0.025	Y
WD2 09	9	154621	234498	99996	1148	4	32.749	0.327	1.310	0.779	Y
WD2 10	10	155176	167218	101850	1151	2	51.903	0.519	1.038	0.887	Y
WD2 11	11	152364	195466	102035	1158	2	36.341	0.363	0.727	1.222	Y
WD2 12	12	153146	155349	101457	1138	3	47.800	0.478	1.434	0.030	Y
WD2 13	13	147466	314428	95620	1160	3	56.224	0.562	1.687	0.026	Y
WD2 14	14	152346	165971	100524	1134	3	23.717	0.237	0.712	0.058	Y
WD2 15	15	151565	156268	98918	1144	3	41.096	0.411	1.233	0.035	Y
WD2 16	16	149500	233193	96986	1129	4	56.328	0.563	2.253	0.460	Y
WD2 17	17	151826	251488	98672	1119	3	56.369	0.564	1.691	0.026	Y
WD2 18	18	152123	125931	97663	1115	4	38.695	0.387	1.548	0.663	Y
WD2 19	19	148275	190044	94286	1128	4	43.066	0.431	1.723	0.598	Y
WD2 20	20	152504	180797	97977	1117	3	56.955	0.570	1.709	0.026	Y
WD2 21	21	147645	277927	97038	1102	3	49.234	0.492	1.477	0.030	Y
WD2 22	22	149368	206623	97361	1112	4	39.001	0.390	1.560	0.658	Y
WD2 23	23	147739	201129	96245	1092	4	54.895	0.549	2.196	0.472	Y
WD2 24	24	149381	276770	97152	1147	3	55.136	0.551	1.654	0.027	Y
WD2 25	48	149182	694049	96685	1093	3	41.943	0.419	1.258	0.035	Y
WD2 26	72	148993	985576	95410	1099	4	48.412	0.484	1.936	0.534	Y
WD2 27	96	151358	1071152	97533	1091	4	20.572	0.206	0.823	1.198	Y
WD2 28	120	148823	1242798	95081	1073	4	52.077	0.521	2.083	0.497	Y
WD2 29	144	151308	1015908	97375	1085	4	50.540	0.505	2.022	0.512	Y
WD2 30	168	152057	895071	97932	1094	3	43.862	0.439	1.316	0.033	Y
WD2 31	192	149679	1583664	95895	1126	4	31.807	0.318	1.272	0.801	Y
WD2 32	360	143410	3329816	93557	1138	4	22.156	0.222	0.886	1.121	Y

Lung Fluid Blanks

Initial Radium [pCi] = 0
Initial Radium [Bq] = 0.000

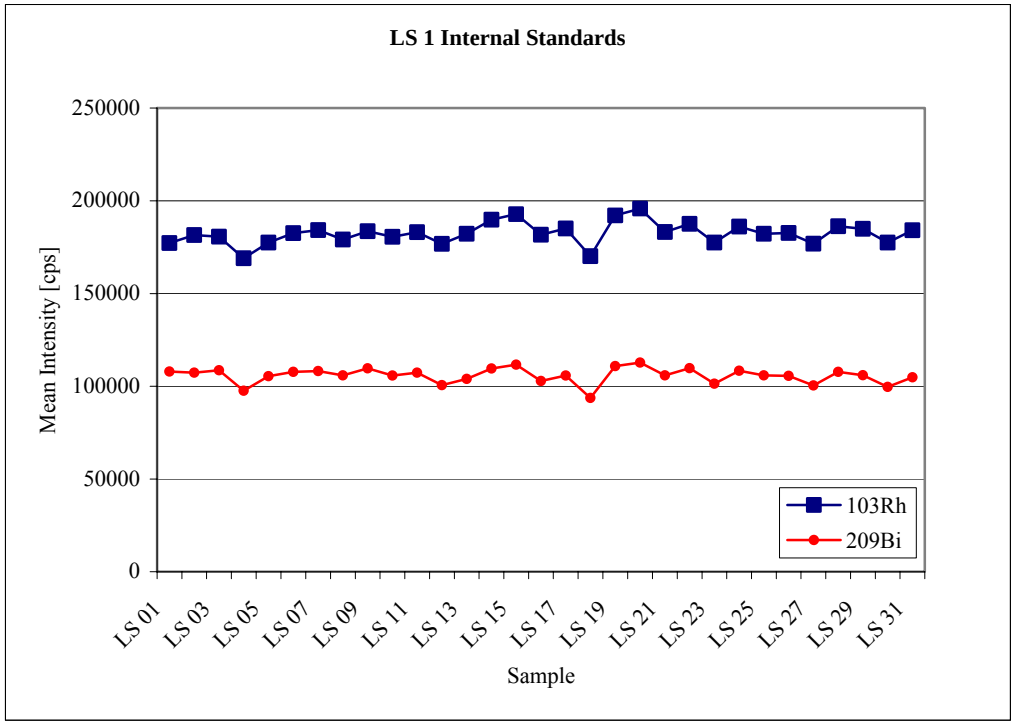
	Mean Intensity [cps]							Weighted Mean and σ Calculation		
In Sample:	^{103}Rh	^{138}Ba	^{209}Bi	^{238}U	^{226}Ra	Intensity % RSD	RSD	σ	w_i	w·M
BL 01	148137	81635	97447	1202	3	46.074	0.461	1.382	0.523414386	1.570243157
BL 02	147222	68041	96759	1181	3	58.228	0.582	1.747	0.327713056	0.983139168
BL 03	143324	69828	91767	1211	4	31.121	0.311	1.245	0.645316739	2.581266954
BL 04	145878	58334	94331	1187	3	32.961	0.330	0.989	1.02271996	3.068159881
BL 05	147280	52557	95504	1179	3	25.568	0.256	0.767	1.69966752	5.09900256
BL 06	148604	49131	96762	1173	2	59.783	0.598	1.196	0.699494975	1.39898995
BL 07	145768	52925	94745	1190	2	62.870	0.629	1.257	0.632489162	1.264978324
BL 08	145457	51855	94743	1189	4	64.680	0.647	2.587	0.149396353	0.597585411
BL 09	145217	50223	94212	1193	3	53.643	0.536	1.609	0.38612807	1.158384211
BL 10	139742	53273	92852	1204	3	36.800	0.368	1.104	0.820468389	2.461405167
BL 11	143456	49146	95691	1197	3	35.355	0.354	1.061	0.888905938	2.666717814
BL 12	138618	57334	90426	1221	3	31.974	0.320	0.959	1.086834834	3.260504501
BL 13	143129	49737	94853	1196	3	33.466	0.335	1.004	0.99208727	2.976261811
BL 14	143207	47051	95214	1178	3	67.748	0.677	2.032	0.242083134	0.726249402
BL 15	143294	45389	95867	1182	3	43.731	0.437	1.312	0.5810034	1.743010199
BL 16	141134	50412	93811	1204	3	48.580	0.486	1.457	0.470806521	1.412419563
BL 17	143090	49480	94283	1195	3	78.242	0.782	2.347	0.181500407	0.544501222
BL 18	141761	48031	94026	1203	3	39.528	0.395	1.186	0.711128049	2.133384147

M_w	2.95545444
σ_{Mw}	0.287942316

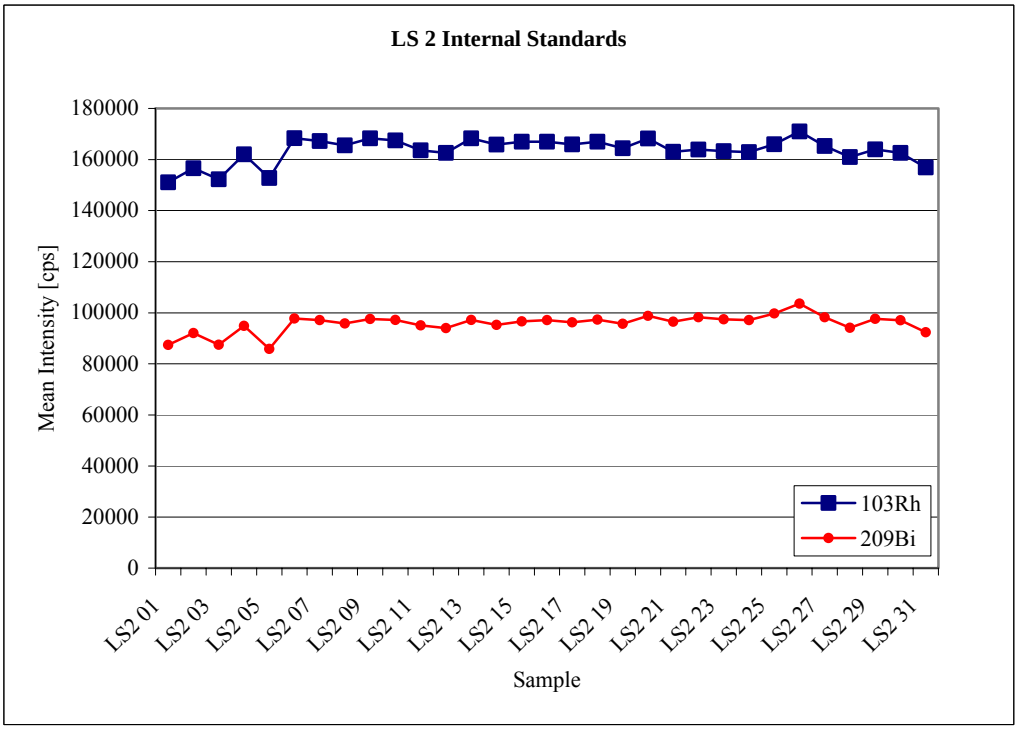
APPENDIX B

INTERNAL STANDARD CONFIRMATION

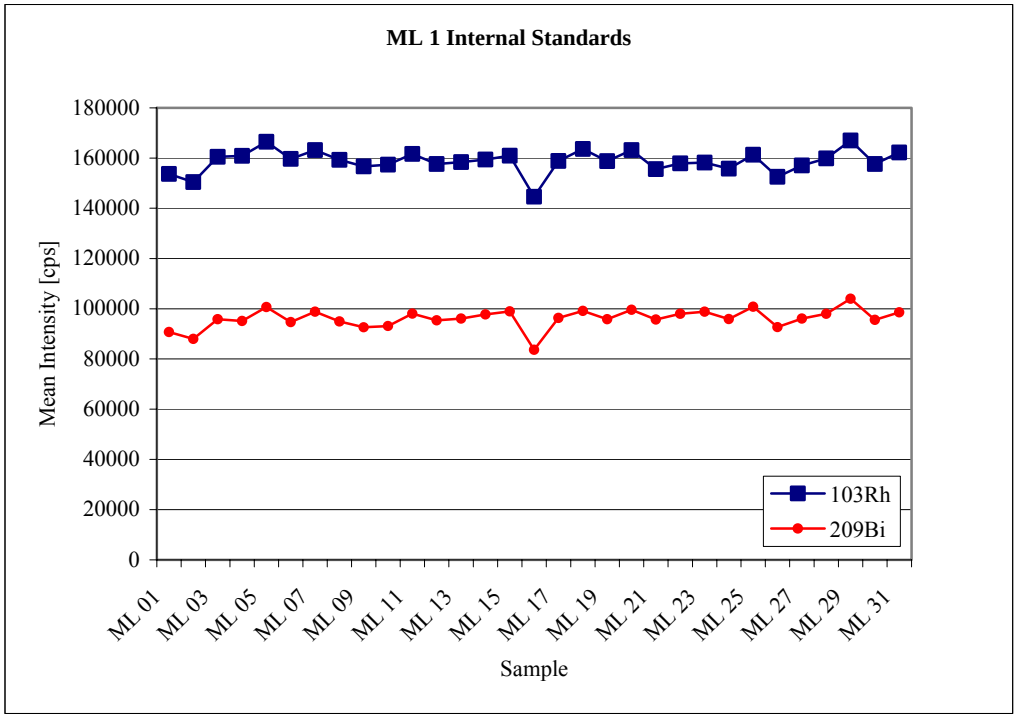
Lake Sand Run 1 Internal Standards



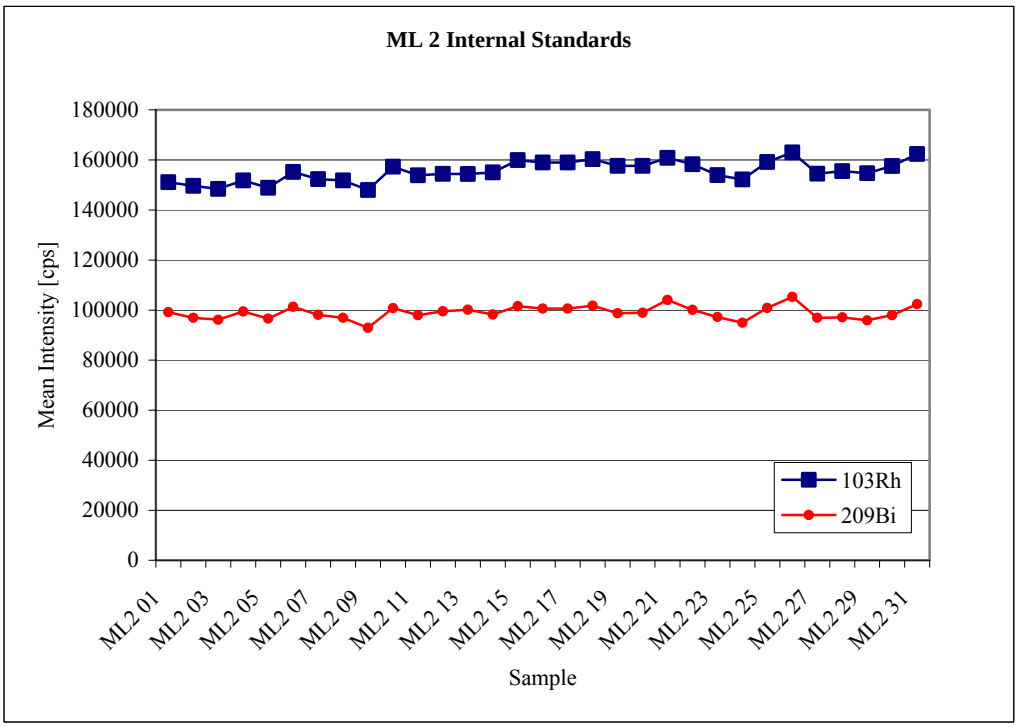
Lake Sand Run 2 Internal Standards



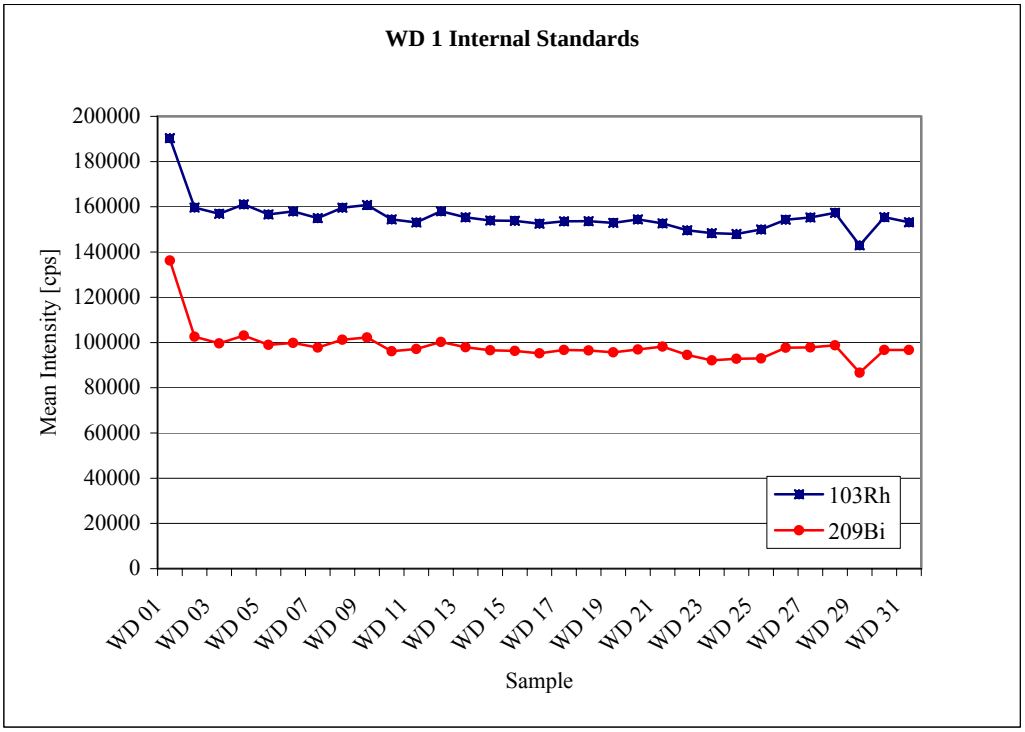
Mud Lake Run 1 Internal Standards



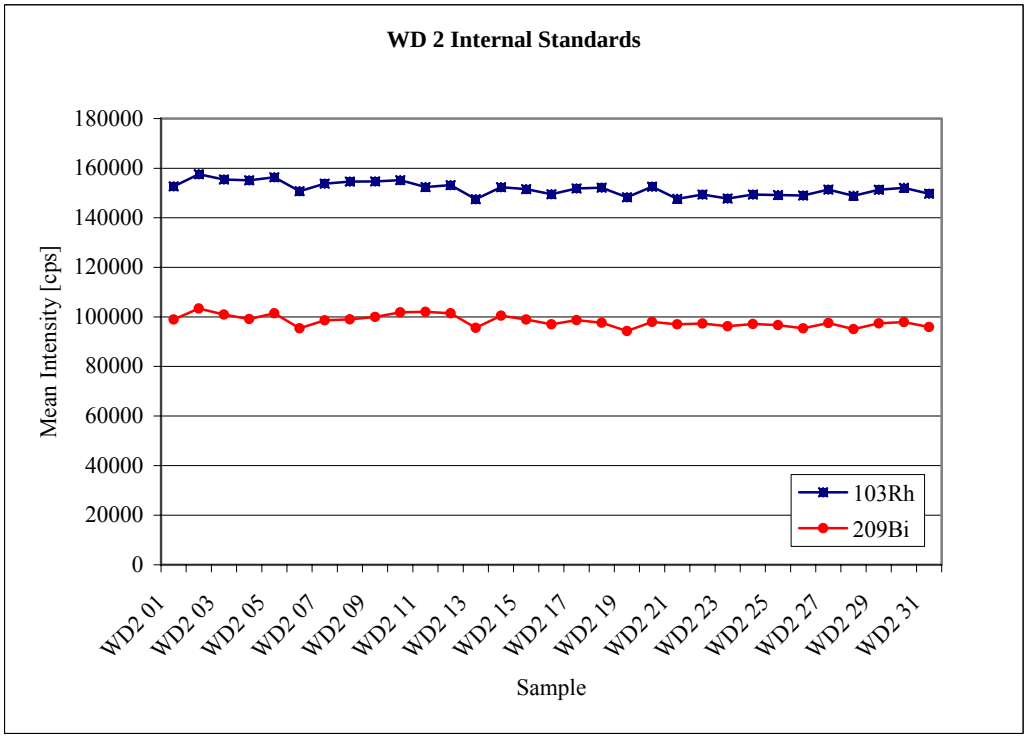
Mud Lake Run 2 Internal Standards



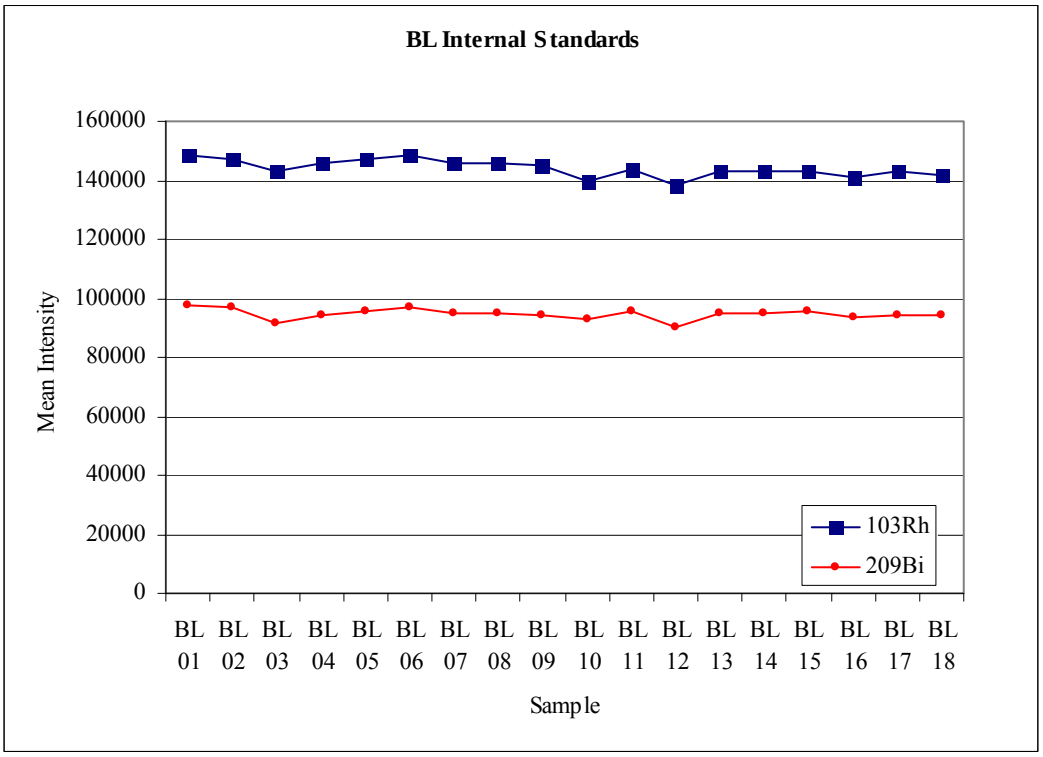
West Delta Run 1 Internal Standards



West Delta Run 2 Internal Standards



Blank (SUF) Internal Standards



APPENDIX C

RADIOANALYTICAL LAB RESULTS

DEPARTMENT OF THE AIR FORCE
AIR FORCE INSTITUTE FOR OPERATIONAL HEALTH (AFMC)
BROOKS AIR FORCE BASE, TEXAS

09 Feb 2004

Radiation Surveillance Division
Radioanalytical Branch
2350 Gillingham Drive
Brooks AFB, TX 78235-5103

Dr. Ian Hamilton
Department of Nuclear Engineering
Texas A&M University
3133 TAMU
College Station, TX 77843-3133

Dear Dr. Hamilton

Attached you find results of the sample analysis and associated quality assurance data for the samples we analyzed for you.

Attachment A contains the sample results and associated instrument calibration data. Attachment B contains the associated quality control sample results that were analyzed with each batch.

Please contact me if we can assist you further. I can be reached at (210) 536-5816 or via email dale.thomas@brooks.af.mil.

DALE D. THOMAS III
Chief, Radioanalytical Branch

Attachments
Sample Results and Calibration Data
Quality Assurance Sample Results

SAMPLE ANALYSIS RESULTS REPORTED ON 09-FEB-2004

AFIERA/SDRR ID: 10300483
Customer Address Code: Q00253C
IERA/SDRH
2350 GILLINGHAM DRIVE
BROOKS AFB TX, 78235-5103
ATTN: RADIATION SAFETY OFFICER

IDENTIFICATION:
Base Sample # CO0300520
Workplace or Site ID: 253 BROOKS AFB
DATE COLLECTED: 02-JUN-2003 RECEIVED: 06-JUN-2003 COMPLETED: 02-DEC-2003
Sample Volume Received: 1980.5 GRAM(s)

EPA CODE N/A ACTINIUM 228		3.9E+02	+/- 5.3E+00	Picocuries / Gram
EPA CODE N/A BISMUTH 212		4.2E+02	+/- 1.7E+01	Picocuries / Gram
EPA CODE N/A LEAD 212		2.3E+02	+/- 1.2E+01	Picocuries / Gram
EPA CODE N/A RADIUM 226		9.1E+02	+/- 1.0E+01	Picocuries / Gram
EPA CODE N/A THORIUM 228		4.4E+01	+/- 2.9E+00	Picocuries / Gram
EPA CODE N/A THORIUM 230		8.1E-01	+/- 1.7E-01	Picocuries / Gram
EPA CODE N/A THORIUM 232		2.3E-01	+/- 9.0E-02	Picocuries / Gram
EPA CODE N/A THORIUM 234	<	1.5E+01		Picocuries / Gram
EPA CODE N/A URANIUM 234		1.5E+02	+/- 4.4E+01	Femtocuries / Gram
EPA CODE N/A URANIUM 235		1.8E+01	+/- 1.6E+01	Femtocuries / Gram
EPA CODE N/A URANIUM 238		1.3E+02	+/- 4.2E+01	Femtocuries / Gram

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COMMENTS:

GENERAL BUCKET #1
QUALITY CONTROL FLAG: GAMMA SPECTROSCOPY RESULTS DO NOT MATCH THE ISOT
THORIUM BY ALPHA SPECTROSCOPY. WE SUSPECT THAT THIS IS ATTRIBUTABLE TO
DEGREE OF INSOLUBILITY OF THE SAMPLE MATRIX. MULTIPLE ATTEMPTS AND VAR
TECHNIQUES FOR DISSOLUTION YIELDED COMPARABLE RESULTS FOR THE
ALPHA SPECTROSCOPY MEASUREMENTS. DUE TO THE NON-DESTRUCTIVE NATURE OF
GAMMA SPECTROSCOPY ANALYSIS, THIS METHOD PROVIDES A MORE QUANTITATIVE
CONCENTRATIONS OF RADIUM-228 AND DECAY PROGENY. THORIUM RESULTS OBTAIN
VIA ALPHA SPECTROSCOPY ARE STILL USEFUL FOR COMPARING THE ISOTOPIC RAT
OF THORIUM PRESENT AND SUBSEQUENTLY NORMALIZING THE DATA TO GAMMA SPEC
DATA TO ESTIMATE THORIUM CONCENTRATION(S).
RECOMMENDATION: RECOMMEND THAT THE ISOTOPIC RATIOS OF THORIUM BE NORMA
TO THE GAMMA SPECTROSCOPY RESULTS FOR ASSESSING ELEMENTAL THORIUM
CONCENTRATION.

RESULTS ACCURATE TO 2 SIGNIFICANT FIGURES.
UNCERTAINTY AT 95% CONFIDENCE LEVEL.

If you have any questions concerning the information provided above, please contact AFIERA/SDRR at DSN 240-2061 or commercially at (210) 536-2061 or call ESOH Service Center at 1 888 232-3764.

Mr. Dale D. Thomas, GS-13
Chief, Radioanalytical Branch

SAMPLE ANALYSIS RESULTS REPORTED ON 09-FEB-2004

AFIERA/SDRR ID: 10300482
Customer Address Code: Q00253C
IERA/SDRH
2350 GILLINGHAM DRIVE
BROOKS AFB TX, 78235-5103
ATTN: RADIATION SAFETY OFFICER

IDENTIFICATION:
Base Sample # C00300519
Workplace or Site ID: 253 BROOKS AFB
DATE COLLECTED: 02-JUN-2003 RECEIVED: 06-JUN-2003 COMPLETED: 02-DEC-2003
Sample Volume Received: 2385.5 GRAM(s)

EPA CODE N/A ACTINIUM 228		7.2E+02 +/-	9.1E+00	Picocuries / Gram
EPA CODE N/A BISMUTH 212		9.3E+02 +/-	3.2E+01	Picocuries / Gram
EPA CODE N/A LEAD 212		5.2E+02 +/-	2.6E+01	Picocuries / Gram
EPA CODE N/A RADIUM 226		1.8E+00 +/-	2.0E-02	Nanocuries / Gram
EPA CODE N/A THORIUM 228		7.3E+01 +/-	4.4E+00	Picocuries / Gram
EPA CODE N/A THORIUM 230		1.6E+00 +/-	2.6E-01	Picocuries / Gram
EPA CODE N/A THORIUM 232		6.6E-01 +/-	1.6E-01	Picocuries / Gram
EPA CODE N/A THORIUM 234	<	2.3E+01		Picocuries / Gram
EPA CODE N/A URANIUM 234		6.0E+01 +/-	2.5E+01	Femtocuries / Gram
EPA CODE N/A URANIUM 235	<	1.6E+01		Femtocuries / Gram
EPA CODE N/A URANIUM 238		2.1E+01 +/-	1.5E+01	Femtocuries / Gram

COMMENTS:

MUD LAKE GENREAL BUCKET
QUALITY CONTROL FLAG: GAMMA SPECTROSCOPY RESULTS DO NOT MATCH THE ISOT
THORIUM BY ALPHA SPECTROSCOPY. WE SUSPECT THAT THIS IS ATTRIBUTABLE TO
DEGREE OF INSOLUBILITY OF THE SAMPLE MATRIX. MULTIPLE ATTEMPTS AND VAR
TECHNIQUES FOR DISSOLUTION YIELDED COMPARABLE RESULTS FOR THE ALPHA
SPECTROSCOPY MEASUREMENTS. DUE TO THE NON-DESTRUCTIVE NATURE OF THE
GAMMA SPECTROSCOPY ANALYSIS, THIS METHOD PROVIDES A MORE QUANTITATIVE
PICTURE OF CONCENTRATIONS OF RADIUM-228 AND DECAY PROGENY. THORIUM
RESULTS OBTAINED VIA ALPHA SPECTROSCOPY ARE STILL USEFUL FOR COMPARING
THE ISOTOPIC RATIOS OF THORIUM PRESENT AND SUBSEQUENTLY NORMALIZING
THE DATA TO GAMMA SPECTROSCOPY DATA TO ESTIMATE THORIUM CONCENTRATION(
RECOMMENDATION: RECOMMEND THAT THE ISOTOPIC RATIOS OF THORIUM BE
NORMALIZED TO THE GAMMA SPECTROSCOPY RESULTS FOR ASSESSING ELEMENTAL
THORIUM CONCENTRATION.

RESULTS ACCURATE TO 2 SIGNIFICANT FIGURES.
UNCERTAINTY AT 95% CONFIDENCE LEVEL.

If you have any questions concerning the information provided above, please contact AFIERA/SDRR at DSN 240-2061 or commercially at (210) 536-2061 or call ESOH Service Center at 1 888 232-3764.

Mr. Dale D. Thomas, GS-13
Chief, Radioanalytical Branch

SAMPLE ANALYSIS RESULTS REPORTED ON 09-FEB-2004

AFIERA/SDRR ID: 10300792
Customer Address Code: Q00253C
IERA/SDRH
2350 GILLINGHAM DRIVE
BROOKS AFB TX, 78235-5103
ATTN: RADIATION SAFETY OFFICER

IDENTIFICATION:
Base Sample # GS0300544
Workplace or Site ID: 253 BROOKS AFB
DATE COLLECTED: 14-AUG-2003 RECEIVED: 14-AUG-2003 COMPLETED: 02-DEC-2003
Sample Volume Received: 256.1 GRAM(s)

EPA CODE N/A ACTINIUM 228		2.2E+00 +/-	2.3E-01	Nanocuries / Gram
EPA CODE N/A BISMUTH 212		9.6E+02 +/-	3.3E+01	Picocuries / Gram
EPA CODE N/A LEAD 212		5.7E+02 +/-	2.9E+01	Picocuries / Gram
EPA CODE N/A RADIUM 226		1.6E+00 +/-	1.0E-02	Nanocuries / Gram
EPA CODE N/A THORIUM 228		4.0E+02 +/-	2.1E+01	Picocuries / Gram
EPA CODE N/A THORIUM 230		7.2E+00 +/-	5.8E-01	Picocuries / Gram
EPA CODE N/A THORIUM 232		2.3E+00 +/-	2.8E-01	Picocuries / Gram
EPA CODE N/A THORIUM 234	<	2.4E+01		Picocuries / Gram
EPA CODE N/A URANIUM 234		4.8E-01 +/-	9.0E-02	Picocuries / Gram
EPA CODE N/A URANIUM 235	<	3.0E-02		Picocuries / Gram
EPA CODE N/A URANIUM 238		2.1E-01 +/-	6.0E-02	Picocuries / Gram

COMMENTS:

WEST DELTA P<105NM/TEXAS A&M
QUALITY CONTROL FLAG: GAMMA SPECTROSCOPY RESULTS DO NOT MATCH THE ISOT
THORIUM BY ALPHA SPECTROSCOPY. WE SUSPECT THAT THIS IS ATTRIBUTALBE TO
HIGH DEGREE OF INSOLUBILITY OF THE SAMPLE MATRIX. MULTIPLE ATTEMPTS
AND VARIED TECHNIQUES FOR DISSOLUTION YIELDED COMPARABLE RESULTS FOR
THE ALPHA SPECTROSCOPY MEASUREMENTS. DUE TO THE NON-DESTRUCTIVE NATURE
GAMMA SPECTROSCOPY ANALYSIS, THIS METHOD PROVIDES A MORE QUANTITATIVE
PICTURE OF CONCENTRATIONS OF RADIUM-228 AND DECAY PROGENY.
THORIUM RESULTS OBTAINED VIA ALPHA SPECTROSCOPY ARE STILL USEFUL
FOR COMPARING THE ISOTOPIC RATIOS OF THORIUM PRESENT AND SUBSEQUENTLY
NORMALIZING THE DATA TO GAMMA SPECTROSCOPY DATA TO ESTIMATE
THORIUM CONCENTRATION(S).
RECOMMENDATION: RECOMMEND THAT THE ISOTOPIC RATIOS OF THORIUM BE
NORMALIZED TO THE GAMMA SPECTROSCOPY RESULTS FOR ASSESSING ELEMENTAL
THORIUM CONCENTRATION.

RESULTS ACCURATE TO 2 SIGNIFICANT FIGURES.
UNCERTAINTY AT 95% CONFIDENCE LEVEL.

If you have any questions concerning the information provided above, please contact AFIERA/SDRR at DSN 240-2061 or commercially at (210) 536-2061 or call ESOH Service Center at 1 888 232-3764.

Mr. Dale D. Thomas, GS-13
Chief, Radioanalytical Branch

VITA

Jason Roderick Cezeaux

20611 Bouganvilla Blossom Trail
Cypress, TX 77433

Education

Master of Science, Health Physics, Texas A&M University, August 2004
Bachelor of Science, Nuclear Engineering, Texas A&M University, May 2003
Minor in Radiological Health Engineering

Professional Experience

Consulting Health Physicist, April 2002 – Present
Foxfire Scientific, College Station, Texas
Graduate Research Assistant, May 2003 – Present
Department of Nuclear Engineering, Texas A&M University
Student Lab Worker, September 2002 – May 2003
Department of Nuclear Engineering, Texas A&M University
Intern, Summer 2000, 2001, 2002
Comanche Peak Steam Electric Station, Texas Utilities, Glen Rose, Texas

Research

"Determination of Petroleum Pipe Scale Solubility in Human Lung Fluid"
Work in Progress Presented: HPS Midyear Meeting, February 2002
"Techniques Employed in Measuring Petroleum Pipe Scale Released by a Dry Rattling Process"
Presented: Waste Management '04, Tucson, Arizona
"In-Situ Spectrographic Analysis of the Vault and Components of a 30-MeV Cyclotron"
Published: Texas A&M University Undergraduate Journal of Science
Presented: ΣΞ International Research Society Forum, November 2002
Presented: National HPS Meeting, June 2002

Professional Development

Nuclear Emergency Planning Course, August 2003
Harvard School of Public Health, Boston, Massachusetts
Radiation Safety Officer Training, May 2003
MFG Environmental, Fort Collins, Colorado
General Plant Information Course, July 2001
Comanche Peak Steam Electric Station, Texas Utilities, Glen Rose, Texas

Accomplishments

National Academy for Nuclear Training Scholar
Institute for Nuclear Power Operations Fellowship
Mensa Member
Eagle Scout with One Bronze Palm