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UNRAVELING ENVIRONMENTAL CONTRIBUTION  
TO FALLOUT FORMATION

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## ABSTRACT

The mechanism of fallout formation remains a poorly-understood phenomenon. Fallout, produced during nuclear detonations, is a mixture of device materials, radioactivity released during fission, and the near surface environment. In this study, the elemental and isotopic composition of fallout is examined to better constrain the environmental contribution to fallout. Nine surface soil samples and six glassy fallout beads, collected from the National Nuclear Security Site (NNSS), were analyzed for major and trace element concentration, mineralogy, and uranium and thorium isotopic composition.

In the Buckboard Mesa the major minerals were found to be quartz, orthoclase, and sanidine. The major elements found in oxide form were silicon, aluminum, and iron. In the Northern Yucca Flat the major minerals were found to be quartz, calcite, calcite magnesian, and dolomite. The major elements found, in oxide form, were silicon, calcium, aluminum, and iron. In the Frenchman Flat the major minerals were found to be quartz, calcite magnesian, and dolomite. The major elements found, in oxide form, were silicon, calcium, and aluminum.

The uranium found in the soil samples is predominantly natural uranium (0.712 -0.771 atom %  $^{235}\text{U}$ ), but two samples showed elevated levels of  $^{235}\text{U}$  (1.44 and 1.60 atom %). The bulk uranium content of the soil was very similar in all locations ranging from 0.521 to 2.980  $\mu\text{g/g}$  with an average of 2.26  $\mu\text{g/g}$ . The bulk thorium content of the soil is highly dependent on location, in the Buckboard Mesa there is  $\sim 22.8$   $\mu\text{g/g}$ , in the Northern Yucca Flats there is  $\sim 2.1$   $\mu\text{g/g}$ , and in the Frenchman Flats there is  $\sim 12.0$   $\mu\text{g/g}$ . The thorium isotopic composition ( $^{232}\text{Th}/^{230}\text{Th}$ ) of the soil also shows locational dependence, in the Buckboard Mesa it is  $8.835 \times 10^4$  to  $1.1349 \times 10^5$ , in the Northern Yucca Flats it is  $2.00 \times 10^5$  to  $2.153 \times 10^5$ , and in the Frenchman Flats it is  $4.917 \times 10^5$  to  $5.460 \times 10^5$ .

The glassy fallout beads are highly enriched in  $^{235}\text{U}$  (79.0-84.2 atom %  $^{235}\text{U}$ ) and  $^{236}\text{U}$  (0.385-0.410 atom %  $^{236}\text{U}$ ). The thorium isotopic composition ( $^{232}\text{Th}/^{230}\text{Th}$ ) of the glassy fallout beads is  $2.330 \times 10^5$  to  $2.733 \times 10^5$ . The beads display remarkable macro-scale isotopic homogeneity. The observed uranium isotopic composition of the glassy fallout beads can be explained by simple two-component mixing between the soil and a device end-member with a nominal isotopic composition of 92 atom %  $^{235}\text{U}$ . Age dating of the glassy fallout beads was attempted using a  $^{234}\text{U}$  to  $^{230}\text{Th}$  decay measurement. Ages of 163 – 213 years were calculated, which proves there is fractionation between the uranium and thorium.

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## INTRODUCTION

The discovery, and later the control, of nuclear fission has undoubtedly changed the world. Scientists understood that harnessing atomic decay could yield vast quantities of energy however it was thought impossible at the time. In 1933 Lord Ernest Rutherford said: “The energy produced by breaking down the atom is a very poor kind of thing. Anyone who expects a source of power from the transformations of these atoms is talking moonshine” [1]. However, the nuclear chain reaction was discovered in 1938 by Otto Hahn of Germany [2]. This led to Enrico Fermi and his team demonstrating that a nuclear fission chain reaction could be controlled, which was proven by the creation of a subcritical graphite pile at the University of Chicago on December 2<sup>nd</sup>, 1942 [3]. This was the first proof that a nuclear power plant could be viable; however, it was a short leap from peaceful nuclear power to nuclear weapons. Leo Szilard was the first to conceive the idea of a nuclear bomb and he patented it with the British government in 1936 [4]. This patent was considered a state secret at the time and not known about until after World War II.

Shortly before the start of World War II, Albert Einstein wrote a letter to President Roosevelt elucidating the dangers that nuclear fission could bring about and that the Germans may currently be pursuing this form of weapons [5]. Roosevelt understood the risk and decided to form a group of scientists to create “The Bomb” for the United States. Thus the Manhattan Project was born.

The Manhattan project, led by J. Robert Oppenheimer, initially created three different nuclear devices: Gadget, Little Boy, and Fat Man. Gadget was the first nuclear device ever detonated; it was an implosion type, fueled with  $^{239}\text{Pu}$ . The scientists were less sure that this style



of device would work and thus they needed to test it. Gadget was used for the, now unclassified, Trinity test [6]. Little Boy was a gun type device fueled with  $^{235}\text{U}$ ; it was dropped on Hiroshima the 6<sup>th</sup> of August, 1945. Fat Man was the same design as Gadget, it was an implosion type device fueled with  $^{239}\text{Pu}$ ; it was dropped on Nagasaki on the 9<sup>th</sup> of August, 1945 [7]. Japan surrendered after the destruction of Hiroshima and Nagasaki and a nuclear device has never again been offensively used.

While America was testing and creating their nuclear arsenal, the rest of the world was busy doing the same; especially the former Soviet Union. Tensions rose between the Soviet Union and the United States during the cold war and only the threat of mutually assured destruction prevented thermonuclear warfare. During the Cold War there were five unclassified incidents in which the US and USSR almost attacked each other [8, 9].

The United States and the Soviet Union understood the dangers of nuclear war and in 1963 they, along with the United Kingdom, signed the Test Ban Treaty. This prevents the testing of nuclear devices above ground, under water, and in outer space [10]. The effects of fallout on the environment were not well understood and there was worry about the level of environmental contamination and the possibility of genetic damage. In 1996 the Comprehensive Test Ban Treaty was brought forth to ban the testing of all nuclear devices regardless of size or location of the test. This treaty has not yet come into effect because all 44 nuclear states must sign and ratify it; 15 states have signed without ratifying (including the USA) and 3 states have neither signed nor ratified it [11].

Beyond limiting the testing of nuclear devices it is important to prevent the spread of nuclear devices. In 1968 The Nuclear Nonproliferation Treaty was opened for signature and currently 190 countries have signed it. At its core the treaty calls for “countries with nuclear

weapons [to] move towards disarmament; countries without nuclear weapons will not acquire them; and all countries can access peaceful nuclear energy” [12]. There are nine states that have nuclear weapons: Britain, France, China, India, Israel, North Korea, Pakistan, Russia, and the United States; however, there may be as many as 20 other nations currently seeking to produce a nuclear weapon [13].

Technical Nuclear Forensics consists of two different parts, the study of pre-detonation events and the study of post-detonation events. Pre-detonation nuclear forensics is about the detection, analysis, and characterization of nuclear and radiological material to prevent proliferation. Post-detonation nuclear forensics is the analysis of fallout debris and the irradiated environment in order to gain knowledge of the device used.

When the Soviet Union dissolved in 1991 much of its nuclear material became unaccounted for. Some of this material has been recovered, but there is still weapons grade material that could be in the hands of terrorists. There is also the possibility of States creating weapons grade material and then selling or giving it to terrorist organizations. This is why there is a strong need to study nuclear forensics. It is imperative to be able to defend the American people against any threat, especially a nuclear based threat.

This study falls in the realm of post-detonation nuclear forensics. Following a near surface nuclear detonation, fallout is formed containing a mixture of the device, radioactivity released by the explosion, and the near ground zero environment. The precise mechanisms of fallout formation remain poorly understood. According to the current model of fallout formation, the nuclear explosion evaporates the surrounding material, including the device. This material quenches to form glasses that trap elements from the detonation and surrounding environment.

Mixing and fractionation behavior may be a function of the different saturated vapor pressures at a given temperature and pressure [14].

## PROCEDURE

Nine surface soil samples (Figure 1) and six glass fallout beads (Figure 2) were collected from three different locations within the National Nuclear Security Site (NNSS) (Figure 3). Three soil samples each were collected from Buckboard Mesa, Frenchman Flat, and Northern Yucca Flat. Samples were collected far enough from the location of any blast such that the soils were minimally contaminated. Collection locations of the glassy fallout beads within the NNSS are unknown.



Figure 1: Surface soil samples inside Teflon beakers.



Figure 2: Glass fallout beads.

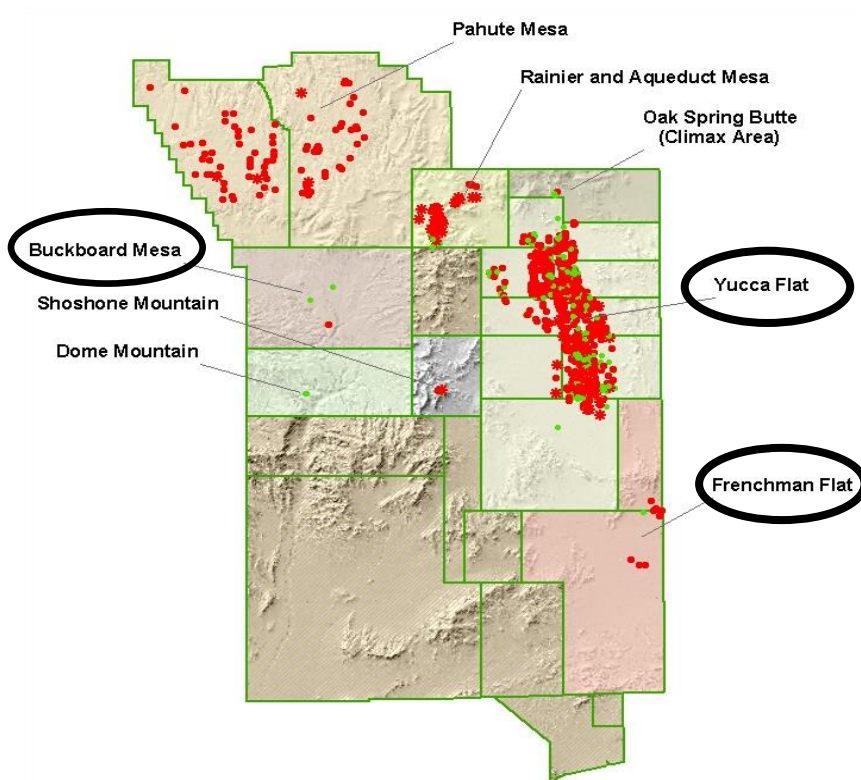


Figure 3: Locations of nuclear detonations in the NNSS. Red indicates sites mapped for surface effects; green indicates that the sites have not been mapped for surface effects [15].

Soil samples were ground into a powder using a clean agate mortar and pestle. A fraction of the powders were separated for bulk X-ray fluorescence and X-ray diffraction analysis. The remaining powder was dissolved in a 2:1 mixture of concentrated  $\text{HNO}_3$  and concentrated  $\text{HF}$ . Glassy fallout beads were dissolved without crushing. New, clean Teflon beakers were used throughout the dissolution procedure.  $\text{HClO}_4$  was used to dissolve the fluorine compounds that formed as a result of the  $\text{HF}$ . These solutions were then dried down, and stock solutions were prepared using 4M  $\text{HCl}$  as shown in Figure 4.



Figure 4: Stock solution of soil in 4M HCl acid.

For uranium concentration and isotopic analysis of the soils and glassy fallout beads aliquots of the stock solutions were transferred to clean Teflon beakers and spiked with  $^{233}\text{U}$ . Spiking a solution with an isotope means adding a known quantity of that isotope dissolved in acid to the aliquot. The  $^{233}\text{U}$  spike used was  $^{233}\text{U}$  Spike 702, which was made by Gary Eppich at Lawrence Livermore National Laboratory (LLNL). This spike is known to have  $1.36023 \times 10^{13} \pm 8.7 \times 10^9$  atoms  $^{233}\text{U}$  per gram of spike.

These aliquots were purified for uranium using ion-exchange chromatography [16]. The samples were dried down and then dissolved them in 1 mL 9M HCl, 25  $\mu\text{L}$   $\text{HNO}_3$ , and 25  $\mu\text{L}$   $\text{H}_3\text{BO}_3$ . The first ion-exchange column was prepared using 1.8 mL of AG1-X8 resin conditioned with 6 mL of 9M HCl. The sample was loaded into the column and then the vial was rinsed twice with 1 mL and 2 mL of 9M HCl to make sure all of the uranium was removed from the Teflon vial. The column was rinsed twice using 2 mL of 9M HCl each time. The uranium was eluted two times using 2 mL of 0.1 M HCl and two times using 3 mL of 0.1 M HCl. The sample was dried down and dissolved in 0.5 mL of 4M  $\text{HNO}_3$  and 25  $\mu\text{L}$  of  $\text{H}_3\text{BO}_3$  for use in the second ion-exchange column.

The second ion-exchange column was prepared using 1 mL of EiChrom U-TEVA resin conditioned with 3 mL of 4M HNO<sub>3</sub>. After loading the sample into the column the vial was rinsed with 0.5 mL and then 1 mL of 4M HNO<sub>3</sub> to be certain that all of the uranium was in the column. The column was rinsed with 2 mL of 4 M HNO<sub>3</sub>, then 0.5 and 1 mL of 9M HCl, and finally with 4 mL of 5M HCl. The uranium was eluted from the column using four steps of 0.5, 1, 2, and 3 mL of 0.1M HCl. The solution was dried down, 3 drops of HNO<sub>3</sub> were added, and dried a final time.

For thorium concentration and isotopic analysis of soils and glassy fallout beads, aliquots of the stock solutions were transferred to clean Teflon beakers and spiked with <sup>229</sup>Th. The <sup>229</sup>Th spike used was <sup>229</sup>Th Spike 5 Alpha, which is made by Gary Eppich at LLNL. This spike is known to have  $1.5.64208 \times 10^{11} \pm 1.37 \times 10^9$  atoms <sup>229</sup>Th per gram of spike.

These aliquots were purified for thorium using ion-exchange chromatography. The thorium samples were run through three different ion-exchange columns. To the sample was added two drops of concentrated HCl, the sample was dried down, then two more drops of concentrated HCl, and then dried down a second time. The sample was then dissolved in 1 mL 9M HCl and 25  $\mu$ L NHO<sub>3</sub>. The ion-exchange column was prepared using 1.8 mL of AG1-X8 resin conditioned with 6 mL of 9M HCl. The sample was loaded into the column, then the vial rinsed twice with 1 mL and 2 mL of 9M HCl to be certain that all of the thorium was removed from the Teflon vial. The column was rinsed twice, using 2 mL of 9M HCl each time. The solution was collected and then dried down.

The second ion-exchange column started by adding two drops of concentrated HNO<sub>3</sub> to the sample, drying the sample down, and then adding two more drops of concentrated HNO<sub>3</sub>, and drying down again. The solution was then dissolved in 1 mL of 8M HNO<sub>3</sub>. The ion-exchange column was prepared using 1.8 mL of AG1-X8 resin conditioned with 6 mL of 8M HNO<sub>3</sub>. The



sample was loaded into the column and the vial was rinsed four times; twice with 1 mL and twice with 2 mL of 8M HNO<sub>3</sub>. This was to ensure that all of the thorium was removed from the vial and in the column. The thorium was then eluted from the column. The first elution used 2mL of 9M HCl. The following elutions used 1, 2, and 3 mL of 0.1M HCl and 0.005M HF. The thorium was then dried down.

The third ion-exchange column was performed the same as the first column.

The samples were analyzed on multi-collector inductively coupled plasma mass spectrometers (MC-ICP-MS) [17] at LLNL. Uranium isotopic composition was measured on a MicroMass IsoProbe MC-ICP-MS. Mass bias and detector gain corrections were made by analyzing the isotopic standard NBL CRM 4321C. Thorium isotopic composition was measured on a Nu Plasma MC-ICP-MS. Mass bias and detector gain corrections were made by analyzing a solution of the isotopic standard U010 spiked with a known concentration of <sup>233</sup>U. Blank measurements were performed prior to each analysis, and blank intensities were subtracted from each analysis. To assess environmental and/or cross-contamination between samples, procedural blanks were also prepared. Procedural blank concentrations were very low, demonstrating that contamination was negligible. No corrections were made for the procedural blank.

Soil powders were analyzed for bulk major and trace element concentration using a Bruker S8 wavelength dispersive X-ray fluorescence spectrometer (WD-XRF) [18]. Samples were analyzed as loose powders in a helium environment. Elemental concentration calculations were performed using the Bruker Quant-Express software. Detection limits for most elements were on the order of 10-100 ppm. Soil powders were also analyzed on a Bruker D8 X-ray diffractometer (XRD) to determine soil mineralogy [19].

## CALCULATIONS

To calculate how much of a given isotope was within the samples isotope dilution mass spectrometry (IDMS) [20] was used. It is assumed that the soils and fallout beads did not have any initial  $^{233}\text{U}$  or  $^{229}\text{Th}$  in them and thus all of the  $^{233}\text{U}$  and  $^{229}\text{Th}$  measured came from the spike. Knowing the mass and concentration of the spike means that the atoms within the spike can be calculated using Equation (1) [20]:

$$n_{spike} = \frac{C_{spike}N_a}{\mathcal{A}_{spike}}, \quad (1)$$

where  $n_{spike}$  is the atoms of the spike added to the aliquot,  $C_{spike}$  is the concentration of the spike (given in ng/g),  $N_a$  is Avogadro's number, and  $\mathcal{A}_{spike}$  is the atomic mass of the spike.

The mass spectrometer gives only the atom ratios of each isotope to each other isotope. The number of atoms of the spike was calculated with Equation (1) and now the number of atoms of each isotope can be calculated. To find the atoms of each isotope Equation (2) [20] is used,

$$n_{isotope} = n_{spike} \left( \frac{n_{isotope}}{n_{spike}} \right), \quad (2)$$

multiplying the number of atoms within the spike by the ratio of atoms of the isotope of interest to that of the spike.

The mass of each isotope within the aliquot is then simple to calculate by multiplying the number of atoms of the isotope by Avogadro's number and dividing by the molecular mass of the isotope. This is shown in Equation (3) [20]:

$$m_{isotope} = \frac{n_{isotope} N_a}{A_{isotope}}. \quad (3)$$

To determine the concentration of the isotope within the original soil the dilution of the sample within the aliquot must be taken into account. The dilution of the sample within the aliquot is defined as the ratio of the mass of the sample to the mass of the aliquot as shown in Equation (4) [20]:

$$D = \frac{m_{sample}}{m_{aliquot}}. \quad (4)$$

Then the mass concentration of each isotope within the original soil is found using the dilution factor determined in Equation (4). The mass concentration of the isotope within the original soil is given by Equation (5) [20]:

$$m_{in\ soil} = \frac{m_{isotope}}{m_{aliquot} D}. \quad (5)$$

To find the total mass of each element (either uranium or thorium) within the original soil the mass concentrations of each isotope of the element are summed as shown in Equation (6):

$$m_{total} = \sum_i m_{in\ soil,i}. \quad (6)$$

The results of these calculations are displayed in Figure 6 through Figure 9.

This is experimentally determined data and as such there is always uncertainty. The initial error comes from two sources, the measurement on the MC-ICP-MS and measuring the masses of the samples. This error must be propagated through the calculations in order to know the validity of the numerical answers. A complete analysis of counting statistics and error propagation can be found in Knoll's textbook, *Radiation Detection and Measurement* [21].

When using Equations (1, 2, 3, 4, 5) the error is propagated as shown in Equation 3.41 of Knoll [21]:

$$\left(\frac{\sigma_u}{u}\right)^2 = \left(\frac{\sigma_x}{x}\right)^2 + \left(\frac{\sigma_y}{y}\right)^2. \quad (7)$$

Equation 6 is purely addition and thus the error is propagated differently, as shown in Equation 3.38 of Knoll [21]:

$$\sigma_u = \sqrt{\sigma_x^2 + \sigma_y^2}. \quad (8)$$

In the uranium decay series,  $^{238}\text{U}$  decays to  $^{234}\text{U}$ , which decays to  $^{230}\text{Th}$ . Measuring the ratio of  $^{234}\text{U}$  to  $^{230}\text{Th}$  allows a model age to be calculated, assuming no initial daughter ( $^{230}\text{Th}$ ) and no addition or removal of  $^{230}\text{Th}$  and  $^{234}\text{U}$  after bead formation. The activity ratio between the daughter and the parent isotope is given by (7) [20]:

$$A = \frac{A_{\text{Th-230}}}{A_{\text{U-230}}} = \frac{N_{230}\lambda_{230}}{N_{234}\lambda_{234}} \quad (9)$$

The age of the fallout beads can then be calculated using (8) [20]:

$$t = \frac{\ln(1-A)}{\lambda_{230}} \quad (10)$$

The result for this calculation is found in Figure 10.

## RESULTS AND DISCUSSION

Using XRD the mineralogy of the nine different NNSS soil samples was determined (Table 1). It was found that soils from the same location generally show similar mineralogy. Buckboard Mesa samples are dominated by silicates, while Frenchman Flat soils contained ~20% carbonates. Northern Yucca Flat samples show more variation between samples, and contain significant amounts of both carbonates and silicates. This is significant because it shows a large amount of mineralogical and chemical heterogeneity in the soil. When determining the environmental contribution to fallout this fact must be taken into account. In the other locations the soil is fairly homogenous and so the elemental concentrations can be average together, however in the Northern Yucca Flat this would not be possible. Figure 5 shows the major elements in oxide form from four samples that were chosen to be representative of their location. Two samples are graphed from the Northern Yucca Flat because its soil was the most diverse. Sample 127, designated as Northern Yucca Flat 1 in Figure 5, was significantly different from samples 132 and 134, designated as Northern Yucca Flat 2 in Figure 5.

Table 1: Mineralogy of the nine soil samples.

<b>Sample ID</b>	<b>Location</b>	<b>Comments</b>	<b>Mineralogy</b>
NTS 86	Buckboard Mesa	Rocky, vegative	Quartz, Orthoclase, Sanidine
NTS 90		Rock sample	Quartz, Orthoclase, Sanidine
NTS 92		Loose rocky	Quartz, Orthoclase, Sanidine
NTS 127	Northern Yucca Flat	Rocky, loose	Quartz, Calcite, Dolomite
NTS 132		Loose soil	Calcite, Dolomite, Ankerite
NTS 134		Very rocky	Calcite Magnesian, Dolomite, Ankerite
NTS 137	Frenchman Flat	Playa	Quartz, Calcite Magnesian, Dolomite
NTS 139		Playa	Quartz, Calcite Magnesian, Dolomite
NTS 141		Loose dirt	Quartz, Calcite Magnesian, Dolomite

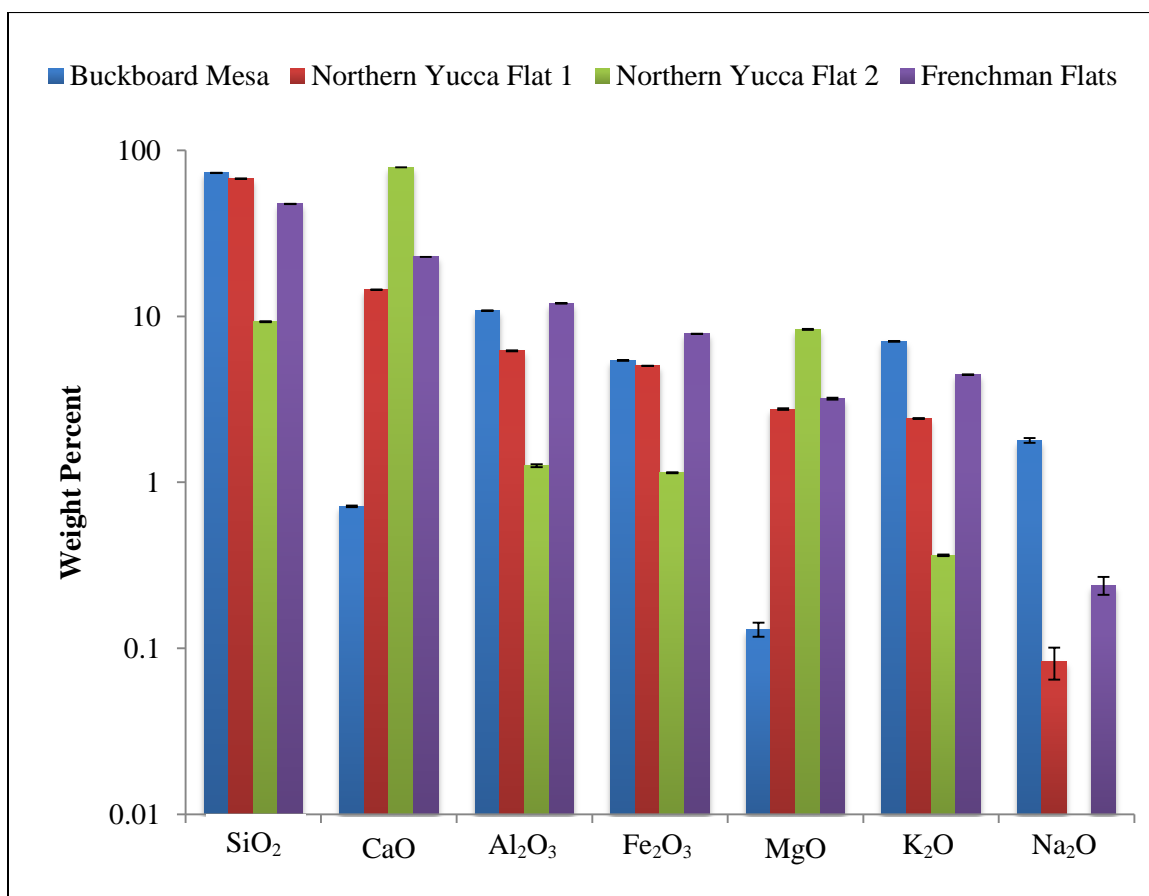


Figure 5: Major elements found in the representative soil samples.

Uranium and thorium concentration results, measured by IDMS, are shown in Figure 6. Uranium concentrations vary by  $<5 \mu\text{g/g}$  within all of the soils, while thorium concentrations are more variable and show grouping based on sample location.



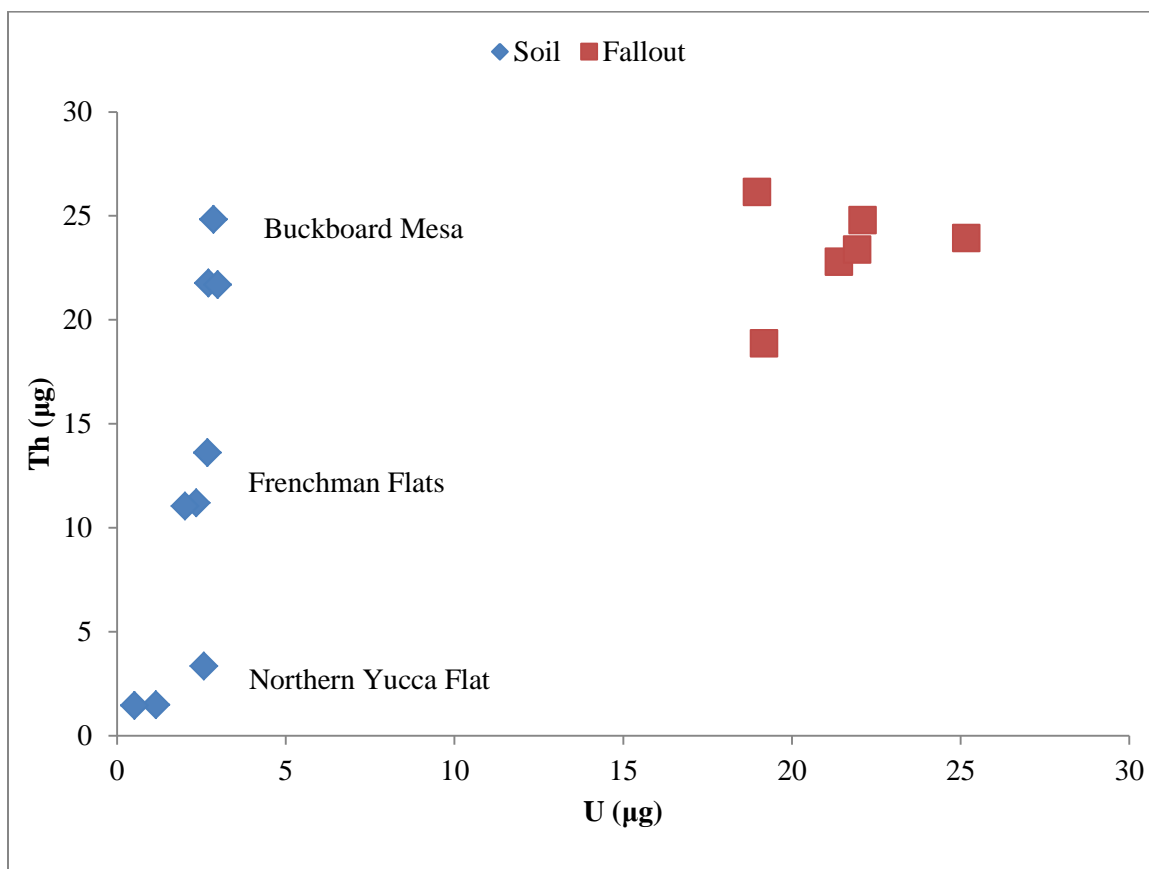


Figure 6: Thorium vs. uranium concentrations in the samples. Error bars are smaller than the symbols.

Most of the natural soil samples collected showed a  $^{238}\text{U}/^{235}\text{U}$  similar to that of natural uranium (Figure 7); however, samples NTS 86 and NTS 132 showed a reduced  $^{238}\text{U}/^{235}\text{U}$  compared to natural uranium. This reduction in  $^{238}\text{U}/^{235}\text{U}$  is significantly above the procedural blank so there is confidence that it does not come from procedural contamination. These samples also have an increased  $^{234}\text{U}/^{235}\text{U}$  and an increased  $^{236}\text{U}/^{235}\text{U}$  (Figure 8) compared to natural uranium. Soil sample NTS 86 falls between the fallout values and natural soil, suggesting that the uranium in this sample may be a mixture of the end-member sources. Soil sample NTS 132 does

not, suggesting that the non-natural uranium in this sample may come from a different source, most likely the detonation of a different device.

Figure 7 shows that the  $^{234}\text{U}/^{235}\text{U}$  is remarkably similar within all of the glassy beads, in contrast with the soils; which display much larger variations. Figure 7 also shows that there are noticeable variations in the  $^{238}\text{U}/^{235}\text{U}$  within the glass beads. This may be a function of where within the cloud the bead formed.

On Figure 7 and Figure 8 a third set of data is plotted and is referred to as the “corrected” fallout. This data arises from trying to correct for the mixing of the natural soil and the device material. An average uranium concentration within the soils of 2.26  $\mu\text{g}$  was used, which does not include the contaminated samples, and assumed equal mass contributions between the environment uranium and the device uranium. Simple subtraction was performed to correct uranium isotopic compositions in the fallout for the environmental contribution. This results in fallout glass enrichments of approximately 92%  $^{235}\text{U}$ .

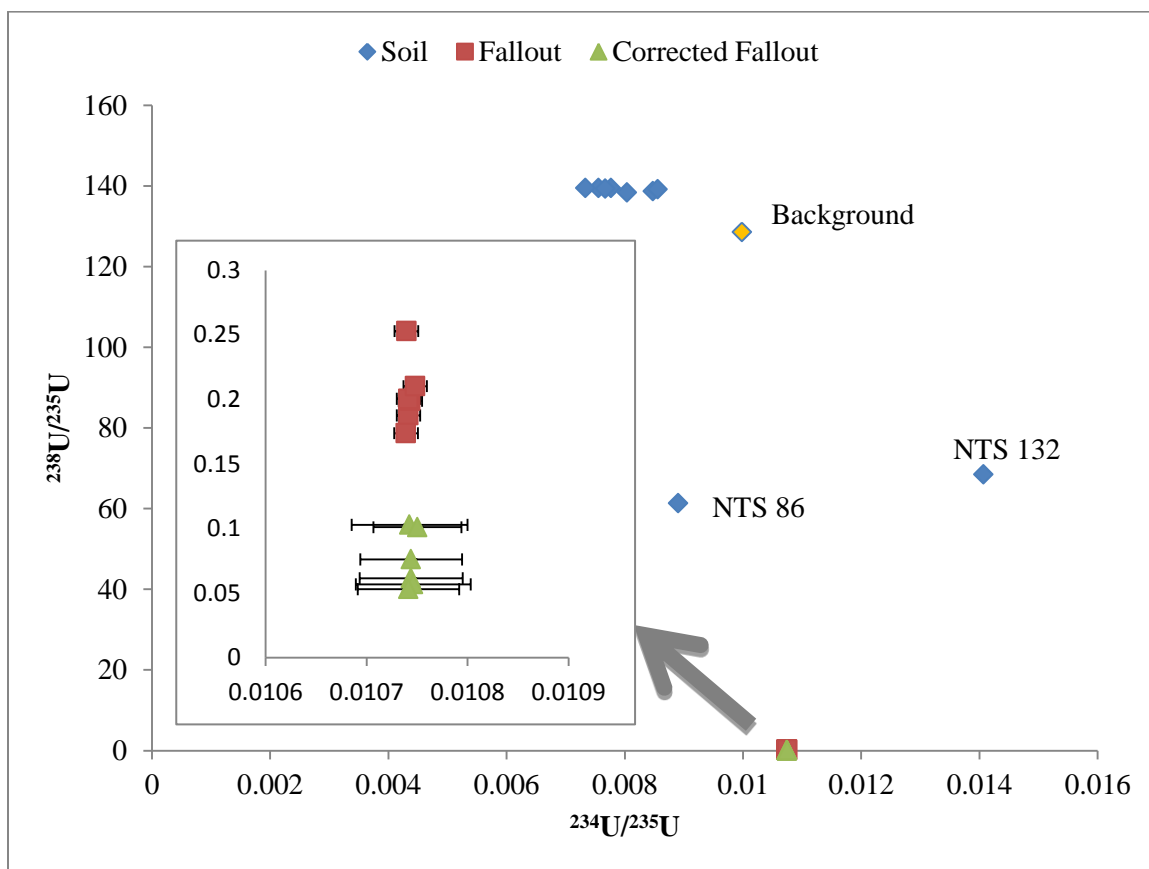


Figure 7: Uranium isotopics normalized to  $^{235}\text{U}$ , an expanded view of the fallout glass is shown.

NTS soil error bars are shown, but are smaller than the symbols.

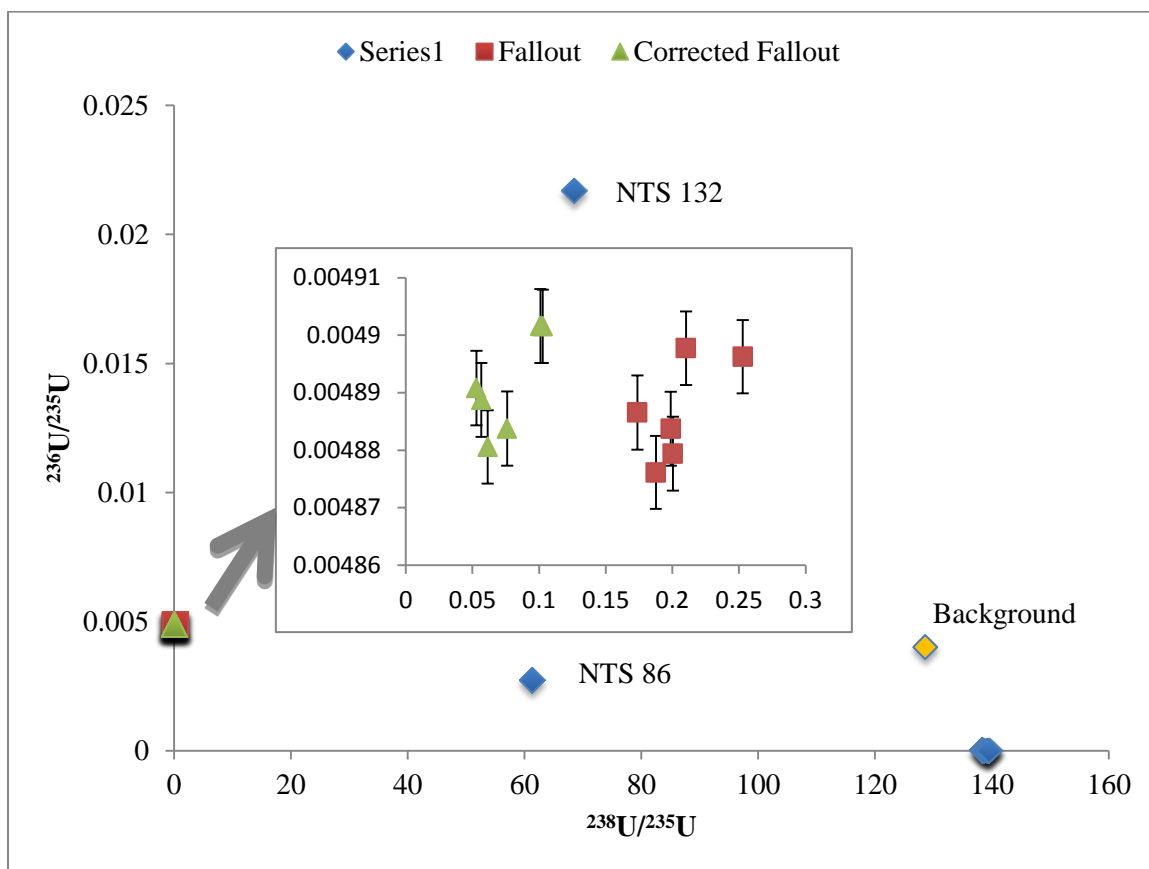


Figure 8: Uranium isotopics normalized to  $^{235}\text{U}$ , an expanded view of the fallout glass is shown.

NTS soil error bars are shown, but are smaller than the symbols.

Strong variance between both the thorium concentrations and the thorium isotopic composition is observed in Figure 9. Soils collected from the three different locations cluster with respect to location, as might be expected, suggesting thorium isotopic composition is characteristic of rock type. Fallout shows less scatter relative to the soil values. This could be due to fallout formation mechanisms or the result of a uniform environmental contribution from a single location. Trying to correct the thorium from the fallout for the environmental contributions

did not provide any useful results. The thorium content is so varied by location one cannot meaningfully define an average thorium content for the NNSS.

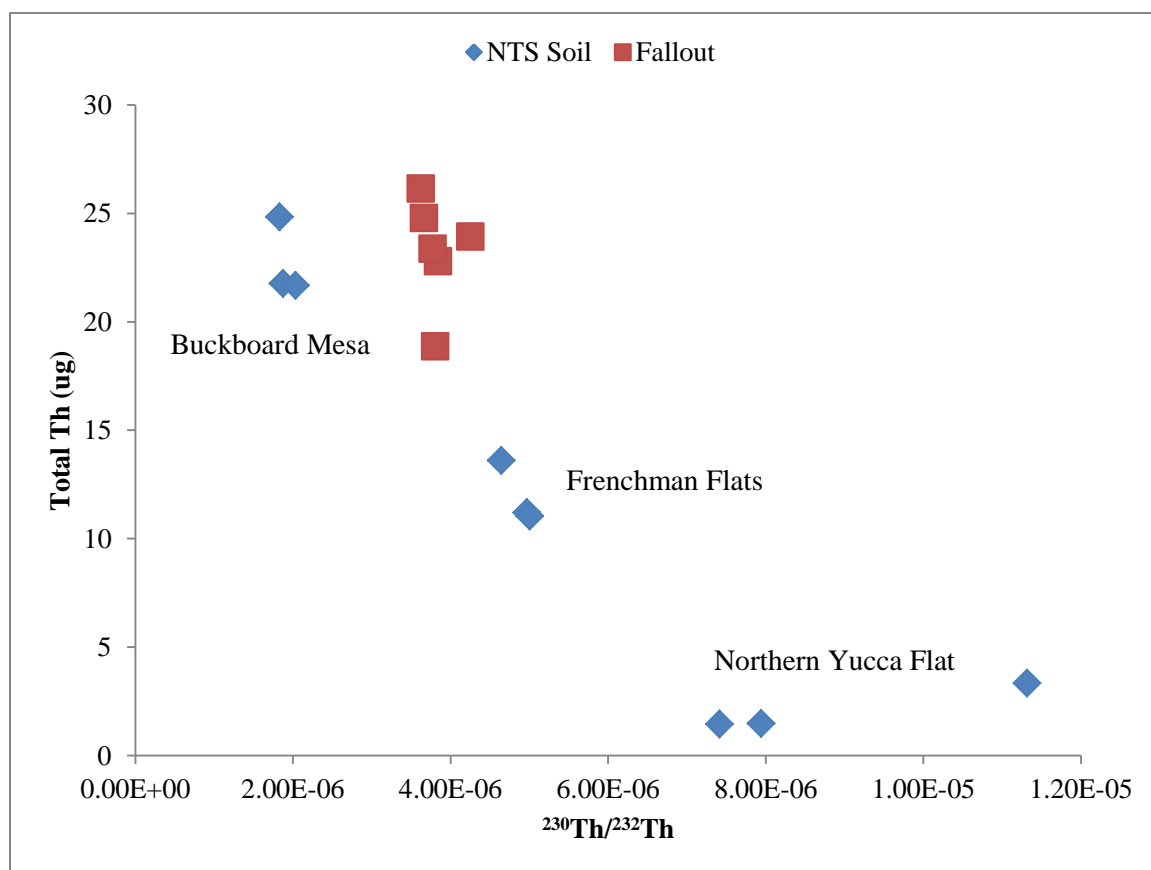


Figure 9: Thorium isotopics vs. thorium concentration. Error bars are shown, but are smaller than the symbols. Grouping is seen based on location of sample.

Using  $^{234}\text{U} - ^{230}\text{Th}$  radio dating, model ages that vary from 163-213 years before present are calculated (Figure 10). These ages cannot be correct as above-ground testing was limited to

the years 1945-1962. Thus, there is excess  $^{230}\text{Th}$  relative to  $^{234}\text{U}$ . To explain this, chemical fractionation between uranium and thorium may have occurred during the formation of the glassy fallout beads. It is unclear whether  $^{230}\text{Th}$  was added or  $^{234}\text{U}$  was lost during this process. Sample 2 had a significantly larger “age” than the rest of the fallout beads. This bead had a  $^{230}\text{Th}/^{234}\text{U}$  ratio approximately 30% higher than the other beads. This is most likely attributed to fractionation between uranium and thorium but why it is different from the other beads is unknown.

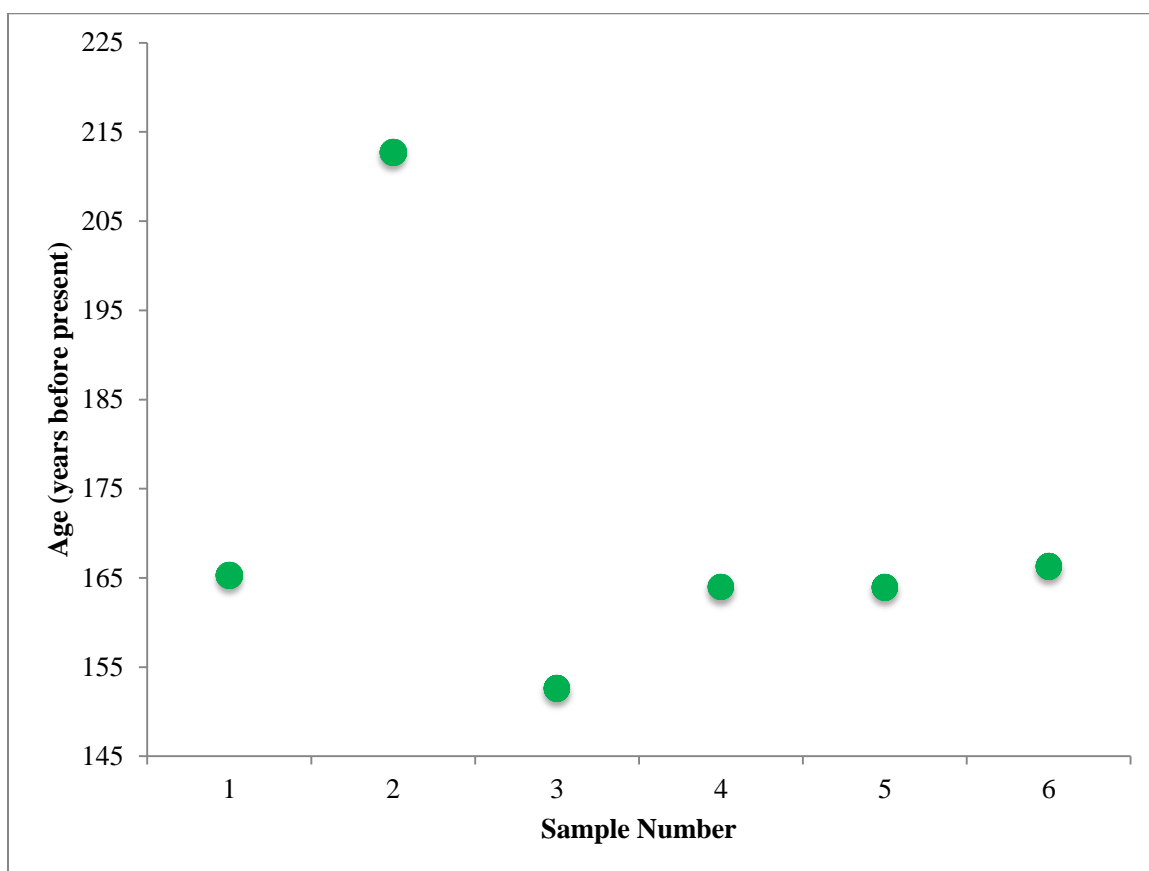


Figure 10:  $^{234}\text{U} - ^{230}\text{Th}$  calculated model age of the glassy fallout bead.

## CONCLUSION

In studying the natural soil of the NNSA it was found to be isotopically, chemically, and mineralogically homogeneous within the Buckboard Mesa and Frenchman Flats. The Northern Yucca Flat is chemically and mineralogically heterogeneous, although isotopically homogeneous. The bulk concentration of uranium within the soils was consistent (less than 5  $\mu\text{g/g}$  spread) and there were strong variations in the bulk concentration of thorium.

After performing the analyses it was found that two of the soil samples contained non-natural uranium isotopic signatures. One looks like it was contaminated by a test with characteristics similar to the investigated glassy fallout beads, the other looks to be contamination of an entirely different composition. This contamination most likely comes from a test that was different than the test which formed the glassy fallout beads.

Bulk analysis of the glassy fallout beads showed, somewhat surprisingly, that they cluster in their isotopic compositions for uranium and thorium. The beads did not, however, cluster in total uranium or thorium content. These differences in total concentration could be a function of the location within the cloud at which the bead is formed.

From these experiments it can be deduced that when the fallout forms the HEU combines with the natural soil, creating a mixture of the two components which can be explained through a simple two component mixing model. There may be thorium contributed from the device or during the formation of fallout glass thorium and uranium are chemically fractionated from one another.

In performing a  $^{234}\text{U}$ - $^{230}\text{Th}$  radio dating, ages for the glassy fallout beads were calculated. The calculated ages were between 163 and 213 years whereas the true age should be between 50-67 years. The incorrect age dating most probably comes from fractionation of the uranium and thorium; however, there is not currently enough data to tell if there is excess thorium or missing uranium.

The fallout beads were all similar in their bulk isotopic compositions. Performing a simple correction for the environmental contributions making simple assumptions, a starting isotopic composition for the fallout of 92%  $^{235}\text{U}$  is calculated. This is consistent with what might be expected. Regrettably, the correctness of the assumptions cannot be verified with the present data.



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## **ACADEMIC VITA**

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### **Education**

B.S., Nuclear Engineering, 2013, The Pennsylvania State University, University Park, PA

### **Honors and Awards**

- PSNES Leadership and Service Award, Penn State Nuclear Engineering Society, March 2013
- Nuclear Forensics Undergraduate Scholarship, South Carolina Universities Research Foundation, April 2012
- Schreyer Honors College, September 2011 to May 2013
- Dean's List 8 of 8 semesters
- Paul Morrow Endowed Scholarship, College of Engineering, April 2009

### **Association Memberships/Activities**

- Institute of Nuclear Material Management
  - Vice-President of PSU Chapter
- American Nuclear Society
- Tau Beta Pi
- Alpha Nu Sigma

## **Research Experience**

### **Nuclear Forensics Intern at Lawrence Livermore National Laboratory**

"Unraveling Environmental Contributions of Fallout Formation"

Mentored by Gary Eppich and Dr. Kim Knight

- Performed sample preparation using a variety of acids, including concentrated Hydrofluoric and Perchloric acid
- Purified aliquots for Uranium and Thorium using ion exchange resin columns
- Calculated the isotopic abundances of Uranium and Thorium within the soil and the fallout, then used these values to determine an age of the samples
- Worked in a Class 100 clean room

### **Research Assistant at the Penn State Intense Laser Laboratory**

Mentored by Dr. Jovanovic

- Worked on multiple projects involving Shaped Femtosecond Laser Pulse Spectroscopy for Nuclear Forensics
- Created the LabView code to automate the equipment and data acquisition, utilizes Genetic Algorithms
- Assembled the optics, laser path, and vacuum chamber

## **Research Interests**

I am interested in the use of radiation detection for homeland security applications, specifically the prevention of the proliferation of special nuclear material using liquid organic scintillators.

## **Professional Presentations**

"Unraveling Environmental Contribution to Fallout Formation". Nuclear Security Policy and Nonproliferation Technology Workshop. Brookhaven National Laboratory, Upton, New York. January 18th, 2013.

"Unraveling Environmental Contribution to Fallout Formation". Nuclear Forensics Undergraduate Scholars Symposium. Los Alamos National Laboratory, Los Alamos, New Mexico. December 17th, 2012.

## **Publications and Papers**

### Journal Papers

P. Ko, K. Hartig, J. McNutt, R. Schur, T. Jacomb-Hood, and I. Jovanovic, "Adaptive femtosecond laser-induced breakdown spectroscopy for nuclear forensics," submitted to Review of Scientific Instruments (2012).

K.C. Hartig, J.P. McNutt, P. Ko, T.W. Jacomb-Hood, and I. Jovanovic, "Pulse Chirp Effects in Ultrafast Laser-Induced Breakdown Spectroscopy," submitted to Journal of Radioanalytical and Nuclear Chemistry (2012).

Conference Proceedings

K. Hartig, J. McNutt, P. Ko, T. Jacomb-Hood, R. Schur, and I. Jovanovic, "Chirped-Pulse Femtosecond Laser-Induced Breakdown Spectroscopy for Nuclear Forensics," 2012 Annual OSA Conference, Rochester, NY, October 14-18, 2012.

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P. Ko, J. McNutt, K. Hartig, R. Schur, T. W. Jacomb-Hood, and I. Jovanovic, "Uranium Characterization by Shaped Femtosecond Laser Induced Breakdown Spectroscopy," submitted to the Institute of Nuclear Materials Management 53th Annual Meeting, Orlando, FL, July 15-19, 2012.