Uranium Uptake for *Capsicum Annuum* in Various Growing Conditions by

Jenelle E. Parson

A THESIS

Submitted to

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Honors Baccalaureate of Science in Nuclear Engineering (Honors Scholar)

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Abstract approved:

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Uranium is a naturally occurring radioactive element that is commonly found in water, soil, and rock. It can also occur in elevated concentrations in ore bearing bodies and be concentrated through human activities. This thesis focuses on uranium uptake for *Capsicum annuum* for three growing conditions. These include a set of plants grown in hydroponic systems and two traditionally grown, one with constant contamination and one with a single acute spike. In addition to the dose scenarios there are two controls, a hydroponic system and a traditionally grown. Mature pepper plants were purchased. Commercially grown plants were cropped to remove mature seed pods; the plants were then allowed to grow (uncontaminated) until new peppers were just forming. At that point, uranium as uranyl acetate was added. Uranyl acetate was dissolved in water to a concentration of 50 μ g/mL for the hydroponic and the constant contamination group. The one-time spike contained the same amount of uranium as received by the total constant contamination traditionally grown plants, but applied in one application such that concentration was 700 g/mL applied in 50 mL. The peppers collected prior and post contamination were analyzed using neutron activation analysis (NAA). Uranium concentration ratios using a plant-to-soil and a plant-to-water ratio were developed for the different growing conditions. While uranium uptake into the pepper plant fruit was comparable for each condition, the uranium uptake into the hydroton for the hydroponic systems was considerably less, yielding a lower concentration ratio when using soil concentrations. When the concentration ratio was calculated using the water uranium concentration, resulting concentration ratios were similar for all growing conditions, but overall higher for traditionally contaminated systems. This was due to overall lower uranium

concentrations in water for traditionally contaminated systems. This suggests that for hydroponic systems, the concentration ratio using soil concentrations may not be appropriate for comparison with traditional systems and it may be more appropriate to use water concentrations.

Key Words: uranium, peppers, radiation

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I understand that my project will become part of the permanent collection of Oregon State University, University Honors College. My signature below authorizes release of my project to any reader upon request. I also affirm that the work represented in this thesis is my own work.

Jenelle E. Parson, Author

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DEDICATION

This work is dedicated to my father, whose memory always inspires me to be a better person.

Uranium Uptake for Capsicum Annuum in Various Growing Conditions

INTRODUCTION

Uranium is a naturally occurring radioactive material (NORM) which is present in rocks, soil, and sea water. Higher concentrations of Uranium occur in uranium ore. There are three naturally existing isotopes of Uranium: U-234 (0.0055%), U-235 (0.720%), and U-238 (99.2745%) (Korea Atomic Energy Research Institute, 2000). Uranium is of interest due to its ability to fission and the large amounts of energy produced during fission, which can be used in a nuclear reactor to produce steam and thus electricity. Of the naturally occurring uranium isotopes, U-235 is of interest in nuclear reactors due to its ability to fission with thermal neutrons. For use in reactors, uranium needs to be transformed into a useful form; this process, also known as the nuclear fuel cycle, is described in the following figure (Argonne National Laboratory, 2011).



Figure 1 Nuclear Fuel Cycle (Argonne National Laboratory, 2011)

There are three main techniques for extracting uranium ore: open pit mining, underground mining, and in-situ techniques (World Nuclear Association, 2011). Open pit mining is used when the uranium

deposits are close to the surface and involves the creation of large pits that encompass the uranium deposit. This technique requires the movement of large amounts of material to access the ore (World Nuclear Association, 2011). Underground mining is utilized when the uranium deposits are not near the surface. While underground mines have the benefit of having little surface disturbance and less removal of material, there is need for ventilation systems to prevent inhalation of uranium (World Nuclear Association, 2011). In-situ leach mining brings uranium to the surface by circulating oxygenated ground water through the orebody to dissolve uranium oxide (World Nuclear Association, 2011). The uranium oxide can be then recovered as a solution.

When the uranium ore is mined it contains a small percentage of uranium, as low as 0.1% uranium (World Nuclear Association, 2011). Due to the low concentration of uranium, the uranium needs to be extracted from the uranium ore through milling. The uranium ore is reduced to powder and uranium is recovered through leaching with a strong acid or alkaline solution, which dissolves the uranium oxide (League of Women Voters Education Fund, 1985). The uranium oxide is precipitated from the solution and dried. The result is a uranium oxide powder, referred to as yellowcake (Figure 2) (Brown, 2012), which contains uranium concentrations of 80% or higher (World Nuclear Association, 2011).



Figure 2 Uranium Yellowcake (Brown, 2012)

After being mined, Uranium needs to be enriched to 3% to 5% U-235 for use in nuclear reactors. This occurs by separation of the U-235 and U-238 based upon molecular weight. The uranium oxide is refined to uranium dioxide and then converted to uranium hexafluoride using hydrogen fluoride (World Nuclear Association, 2011). During enrichment, gaseous uranium hexafluoride is passed through a centrifuge which uses mass difference to separate U-235 from U-238. At this point, uranium hexafluoride is converted back to uranium oxide (World Nuclear Association, 2011). Once enriched, reactor fuel is usually fabricated into ceramic pellets and loaded into fuel rods to be used in the reactor. Fuel rods are then put into assemblies which will remain in the reactor until it is no longer economical. After being removed from the reactor, the spent fuel is stored in spent fuel pools for at least five years to allow for the decay of shorter-lived fission products (United States Nuclear Regulatory Commission, 2012). As the spent fuel pool is filled with newer fuel, older fuel is transferred to dry cask storage.

During the various steps in the nuclear fuel cycle, there is waste produced that contains uranium and other radioactive materials (League of Women Voters Education Fund, 1985). During the mining and milling processes, waste is created through the leaching process. When the solution is added to the ore, there will be waste left behind which will contain un-dissolved uranium and its progeny. This waste, referred to as uranium mill tailings, will contain the bulk of the radioactivity of the ore (League of Women Voters Education Fund, 1985). Tailings are a sand-like material that contains radioactive material such as radium, selenium, molybdenum, uranium, and thorium (U.S. Environmental Protection Agency, 2011). Often these tailings are left in mined out pits; an image of uranium mill tailings at the Atlas site in Moab, Utah is shown in Figure 3 (United States Nuclear Regulatory Commission, 2012).



Figure 3 Uranium Mill Tailings at the Altas Site in Moab, Utah (United States Nuclear Regulatory Commission, 2012)

In 1983, it was estimated that uranium mill tailings in the United States occupy a volume of 96,500,000 m³, which is the largest volume of radioactive waste in the country (League of Women Voters Education Fund, 1985). A map of tailing locations is seen in Figure 4 (U.S. Environmental Protection Agency, 2011). Prior to 1970, tailing piles were neglected, since their radioactivity was viewed as relatively small; as a result, uranium mill tailings were left abandoned and unprotected (League of Women Voters Education Fund, 1985). It wasn't until 1978, with the Uranium Mill Tailings Act, that mill tailings management was required (League of Women Voters Education Fund, 1985). It wasn't until 1978, the tailings can also be an external hazard to people near the tailings via gamma radiation. Additionally, the tailings can leach into a nearby water source that may be used for drinking water.



Figure 4 Locations of Uranium Mill Tailings (U.S. Environmental Protection Agency, 2011)

Uranium containing waste is produced similarly during the enrichment process. As uranium hexafluoride is enriched to a sufficient U-235 content, there will be a waste stream that is depleted of U-235. Waste is stored either as gaseous uranium hexafluoride or U_3O_8 . Uranium hexafluoride is a highly corrosive material when in combination with water.

Uranium Movement in the Environment

Once in the environment, the mobility of uranium depends on the properties of the soil as well as the uranium species (Roivainen, et al., 2011). The most important factor in the mobility of uranium in soil is pH, while clay and soil content had little effect (Roivainen, et al., 2011).

When soil is contaminated with a radionuclide and a plant is growing there, a portion of the radioactive material will be absorbed into the plant (International Atomic Energy Agency, 2012). The transfer from soil to plant can vary depending on the radionuclide involved, soil properties, and the type of crop (Roivainen, et al., 2011). To describe how much of the radionuclide will transport, concentration ratios (CR) are calculated by comparing the dry weight concentration of the plant to the dry weight concentration of soil (International Atomic Energy Agency, 2010), Equation 1. The use of dry weights is to reduce uncertainty in measurements (International Atomic Energy Agency, 2010).

$$CR = \frac{Dry \, Weight \, Concentration \, in \, Plant}{Dry \, Weight \, Concentration \, in \, Soil}$$
 Equation 1

For uranium, information has been tabulated for the following plants: vegetation cereals, maize, leafy vegetables, non-leafy vegetables, leguminous vegetables, root crops, tubers, herbs, grasses, pasture, and leguminous fodder. Within these types of plants, different soil types have been tabulated; these include sand, loam, clay, and organic. In this thesis, uranium uptake in pepper plants is investigated. Concentration ratios for fruit of non-leafy vegetables are given in the following table (International Atomic Energy Agency, 2010).

Soil Type	Number of Samples	CR Mean Value	CR Minimum	CR Maximum
Sand	7	1.9E-2	1.3E-3	1.6E-1
Loam	4	2.3E-2	7.6E-3	4.7E-2
Clay	7	1.8E-2	5.0E-3	2.0E-1
All	38	1.5E-2	5.2E-4	2.0E-1

Table 1 Concentration Ratios for Fruit of Non-Leafy Vegetables for Uranium (International Atomic Energy Agency, 2010)

As seen by Table 1, the studies of uranium in soil systems are limited. Additionally due to the nature of hydroponic systems, it may not be appropriate to compare hydroton to soil. For such systems, it may be more appropriate to compare concentration in fruiting body to concentration in water. A similar analysis is performed for aquatic systems, such that (International Atomic Energy Agency, 2010):

$$CR = \frac{Dry \ Weight \ Concentration \ in \ Plant}{Concentration \ in \ Water \ Applied}$$
Equation 2

Neutron Activation Analysis

Since Uranium-238 is an alpha emitter, it cannot easily be measured using conventional β , γ detectors. Instead, a technique such as neutron activation analysis needs to be performed. Neutron activation analysis is an analytical technique is used for measurement of elements in samples and has sensitivities on the parts-per-billion scale or better. The analysis utilizes neutron interactions with the sample to measure elemental concentrations within the sample. The main interaction used is the neutron capture interaction, which is described through the figure below (Glascock, 2012). This interaction creates beta and delayed neutrons through the decay of the compound nucleus, which can be detected easily using conventional β , γ detectors, such as a High Purity Germanium (HPGe) detector.



Figure 5 Neutron Capture Interaction used in NAA (Glascock, 2012)

For uranium, the following interaction is of interest (Mahlman & Leddicotte, 1955):

$${}^{238}_{92}U(n,\gamma){}^{239}_{92}U \xrightarrow{\beta_{-}}{239}_{93}Np \xrightarrow{\beta_{-}}{239}_{94}Pu \xrightarrow{\alpha}{235}_{92}U$$
Equation 3

In this decay chain, the radionuclides U-239 and Np-239 emit both gammas and betas as they decay. The half-lives of these radionuclides are displayed in Table 2. As seen by the table the half-life of U-239 is very short, making it unreasonable to measure due to a need for sample cool down after irradiation to allow for decay of short-lived fission products. Similarly, due to the long half-life of Pu-239 it will take a long time for sufficient decay to be measured. Thus it's more desirable to measure Np-239, which has a half-life of 2.36 days.

Radionuclide	Half-life
U-239	23.45 min
Np-239	2.3565 d
Pu-239	24110 y

Table 2 Half-lives of Radionuclides in Uranium Activation Chain (Korea Atomic Energy Research Institute, 1992)

The decay scheme of Np-239 is shown in the figure below (Korea Atomic Energy Research Institute, 1992). As seen by the figure, the decay scheme is very complicated. In this scheme, the two emitted photons of interest are 106.123 keV (27.2%) and 277.599 keV (14.38%) (Korea Atomic Energy Research Institute, 1992).



Figure 6 Decay Scheme of Np-239 (Korea Atomic Energy Research Institute, 1992)

As the sample is present in the reactor, there will be a decrease in uranium atoms due to absorption of neutrons and being fissioned and a proportional increase in daughter atoms. The neutron activation analysis technique in this thesis utilizes the comparative method of analysis. In the sample batch, there are also present standard reference material (SRM) and check standards. The standard reference material contains a known mass of uranium. After a given irradiation period, there will be a resultant count rate. This count rate can be compared to the weight of uranium in the SRM, and through which the activity of the unknowns can be calculated through the following equation (Mahlman & Leddicotte, 1955):

$$Weight of U in sample = \frac{(Counts of Np239 - Background Counts)_{sample}}{\left(\frac{Counts of Np239 - Background Counts}{Weight U}\right)_{standard}}$$
Equation 4

Check Standards are treated as unknowns and the calculated activity is used to assess the accuracy of the results.

METHOD

The goal of this study was to determine the uptake of uranium into pepper plants in different growing conditions and assess different methods of comparison. The overall strategy involved measuring uranium content in pepper plats and soil before and after contamination. These were compared to each other as well as compared to the concentration of uranium in water added.

For this experiment, uranium uptake by pepper plants was assessed over three treatments groups corresponding to a one-time uranium contamination group; a constant uranium contamination group; and a hydroponic uranium group. There were two control groups; one traditionally grown and the other hydroponically grown. Each contamination group contained four pepper plants, while each control group contained two pepper plants. It should be noted that only three pepper plants of the one-time contamination group blossomed, and only one pepper plant of each control survived. The one-time contamination plants were dosed with a one-time application of 50 mL with a concentration of 700 µg/mL of uranyl acetate. The constant contamination group was watered every other day with 50 mL of solution with a concentration of 50 µg/mL uranyl acetate; while the hydroponic group had a contamination group watering level would mirror the amount of water used by the hydroponic plants. However, after one week of the experiment the watering levels were not sufficient due to increased plant growth. Thus, water was supplemented with non-contamination conditions consistent over all constant contamination plants, since they were all in different stages of development.

This experiment took place at the Nuclear Engineering and Radiation Health Physics (NERHP) Greenhouse at Oregon State University (Figure 7). This greenhouse was chosen due to the use of radioactive materials within this experiment. The greenhouse has the benefits of being temperature controlled, and having artificial grow lighting.



Figure 7 NERHP Greenhouse

Temperature was kept around 25 °C, through the use of heaters and a fan system that activated at 30 °C. Since this experiment occurred during winter, artificial lighting was used in the greenhouse from 10 am to 8 pm to supplement natural light.

Determination of Contamination Conditions

For this experiment, uranyl acetate dihydrate (UO₂(CH₃COO)₂·2H₂O) was used. Uranyl acetate was chosen because of its availability and solubility in water. Although uranyl acetate is relatively insoluble in water, it will dissolve up to 8 grams per 100 ml of water (International Bio-Analytical Industries, Inc.). A contamination concentration of 50 µg/mL was chosen for the traditional contamination and the hydroponic system. This concentration was measurable using neutron activation analysis in previous studies at OSU (AlZahrani, 2010). Additionally this concentration provided a conservatively smaller mass than the Radiation Use Authorization (RUA), which specifies the maximum amount of activity that can be used by the authorized user. Due to the large volume of the reservoir and the ambiguity in the water use by the plants in the hydroponic system, it was necessary to be conservative. The one-time contamination concentration was taken as an integral value of the constant contamination for a 28 day study. Using 14 contamination days, this came to a total of 35,000 µg per pepper or 700 µg/mL in a 50 mL application. This is the largest concentration used, which is well below the solubility limit for uranyl acetate.

Timeline of Events

Thirty ornamental pepper plants were bought from Home Depot on January 9, 2012. When bought, several pepper plants were frost-bitten. Over the next month the pepper plants were taken care of and 13 began to flower on January 26th. Of the 30 bought, 15 survived to be used in the experiment. The setup for the experiment was on one side of the greenhouse, as seen in Figure 8. This photograph was taken prior to transferring plants to hydroponic systems. The plants to be contaminated are on the far end of the building, while the closest set of peppers and hydroponic are controls. It was desired to separate the contaminated and control peppers to prevent cross contamination. In the photograph, the heaters, natural light lamps, and fan can also be seen.



Figure 8 Set-up of the Greenhouse

Prior to contamination pepper plants were photographed (APPENDIX A) and peppers were clipped off for analysis on the February 5th and 6th. The peppers were placed in plastic bags. Soil was also taken from each pepper plant and stored in a plastic bag for further analysis. On February 10th, the soil samples were placed in the drying oven at 60 °C and allowed to dry for two days. Once the soil samples were removed, the pepper samples were placed in the oven for two days at 60 °C and allowed to dry. After removing the samples from the oven, they were placed in plastic Ziploc bags until grinded later. It is necessary to dry the samples such that the mass doesn't change during analysis from water evaporating. Additionally drying samples prior to grinding makes it easier to homogenize the sample.

The hydroponic group of peppers was transferred to the hydroponic system on February 14th. A more detailed description of the transferring process is in the next section "Transferring Plants to Hydroponic Systems". The hydroponic control group could not be transferred until February 21st due to a malfunctioning AC Adapter for the vortex. Contamination began on February 15th.

On February 20th, it was noticed that one of the pepper plants was infested with aphids, Figure 9. Pesticide was created such that there were 7 mL per 24 fl oz. The pesticide was applied to affected plant. More infected pepper plants were noticed on March 5th and once-a-day application of pesticide began. However, due to catching these problems hydroponic controls, Control A1 and A2, did not fruit and could not be analyzed in this study. As a result one of the traditionally grown controls, Control B3, was transferred to the hydroponic system. In the rest of this thesis Control B2 will be known as traditionally contaminated control (CT), while Control B3 will be known as hydroponic control (CH)



Figure 9 Pepper Plant Infested with Aphids

The last contamination was on March 20th, when enough peppers developed on each healthy plant for analysis. Peppers plants were photographed again (APPENDIX A) were clipped and soil samples were taken on March 22nd. These were placed in Ziploc bags until drying could occur. Drying of both plants and soil samples occurred on April 4th, and samples were dried for two days. Prior to being placed in the oven it was not noticed that the oven was at a higher temperature corresponding to 100 °C. As a result, the samples were a little cooked, but suitable for analysis.

After being dried, aliquots of the soil samples were encapsulated between April 23rd and 30th. The encapsulation process involved filling 2/5 plastic dram vials with the specific weight of sample. The extruding parts of the vial (i.e. hinge and lip) were then clipped off and the lid of the vial was sealed shut using a soldering iron. The 2/5 dram vial was then sealed in a larger 4 dram vial, which acts as secondary containment. The 4 dram vial was sealed using the same procedure. The soil sample NAA group contains the soil samples as well as three standard reference materials and two check standards. The target mass of soil used was approximately 250 mg, while the standards were approximately 200 g. The standard reference materials used were three replicates of coal fly ash, NIST 1633a; while New Ohio Red Clay

(NORC) and coal fly ash (NIST 1633b) were used for check standards. Since the standards have a greater density than the soil samples, they were mixed with corn starch after being weighed such that they had approximately the same geometry as the soil samples. The hydroton was ground by hand on April 27th and encapsulated the following Monday. The samples were submitted for neutron activation analysis on May 2nd. Samples were irradiated for 14 hours in the Lazy Susan rotating rack facility of the OSU TRIGA Reactor. After allowing short-lived fission products to decay, samples were available on May 10th for analysis. The samples were analyzed by Dr. Leah Minc, the Neutron Activation Analysis Manager at the Radiation Center and Associate Professor in Anthropology at Oregon State University; the results of which are presented in the next section.

Pepper samples were ground using a SPEX Sample Prep Mixer/Mill 8000M on April 21st and 22nd. The samples were ground for two minutes each. Non-contaminated samples were ground first and the mill jars were washed between each use. Samples were opened under the hood and transferred to liquid scintillation vials. The SPEX Mill grinds the sample using a grinding vial set. For this application, the methacrylate grinding vial set was used, Figure 10. This set is composed of a 55 mL vial, aluminum screw on cap, four 9.5 mm methacrylate balls, and two 12.7 m methacrylate balls (SPEX CertiPrep Group L.L.C., 2010). The vial is rotated within the SPEX Mill, causing the methacrylate balls to move inside of the vial, grinding the sample.



Figure 10 Mixer Mill Jar

After grinding all the samples, they were placed in the oven in the liquid scintillation vials with lid removed at 100 °C overnight to remove any excess water. The samples were encapsulated using the same procedure as the soil sample NAA group between the April 27th and May 7th. The standard reference material used for the peppers was three replicates of the liquid standards, while the check standards used were a liquid standard and spinach leaves, NIST 1570. The desired mass of pepper samples was between 700 and 800 mg, however, due to limited pepper production several samples were between 200 and 700 mg. The desired mass for the liquid standards was approximately 200 mg, while the desired mass of the spinach leaves was approximately 100 mg. Due to the low density of the pepper samples and high mass desired, it was necessary to pack the pepper samples to fit in the 2/5 dram vials. The spinach leaf sample was prepared similarly to the soil reference materials. The liquid standard preparation was unique to the other sample types and is described in the following paragraph.

Liquid standards were prepared using ash-less filter paper and a 20 μ g/mL uranyl acetate stock solution. First, the ash-less filter paper was cut into quarters. This was folded such that it fit inside of the 2/5 dram vial approximately the same height as the ground pepper samples, to provide the same geometry. Approximately 200 mg of the stock solution was pipetted onto the paper. The liquid standards were then allowed to dry in the oven for 24 hours at 60 °C. The pepper samples were submitted for NAA on May 8th and irradiated for 21 hours in the Lazy Susan facility of the OSTR; they were available for analysis on May 17th. The analysis for these samples was also performed by Dr. Minc.

Transferring Plants to Hydroponic Systems

The hydroponic system used was the RainForest 66 (RF66) General Hydroponics system. The RF66 system consists of a reservoir, six growing areas, vortex sprayer, and nutrient system, as seen in Figure 11. The reservoir holds a total of 64.352 liters with dimensions of 38.1 cm by 38.1 cm by 38.1 cm, while each growing area is 15.24 cm in diameter (General Hydroponics, 2012). There is the ability for a plant to grow in each of the lid inserts.





Figure 11 RainForest 66 Hydroponic System

Transferring the plants to the hydroponic systems was a several step process. In the hydroponic system, the plants roots rest in hydroton (a soilless growing media) that is contained in CocoTek Liners inside of plastic lids; while the plants are watered at the root level with the vortex sprayer.

Hydroton consists of clay balls that are approximately 1 cm in diameter. The hydroton clay is shaped into balls and fired at around 650 °C (General Hydroponics, 2012). As the clay is fired, it expands creating pockets of air in the pebbles; this results in increased water flow, while the roots are still oxygenated

(General Hydroponics, 2012). The CocoTek liners are made of coconut pith and fibers; they are designed to prevent media from escaping (General Hydroponics, 2012).

Prior to transferring the plants, hydroton was rinsed three times to remove any clay powder on the hydroton. Additionally, the water to be used in the reservoir was created. In the hydroponic system water provides the essential nutrients for the plants through a combination of three products: FloraGro, FloraMicro, and FloraBloom. Concentrations for these products were chosen corresponding suggested concentrations for the blooming and ripening stage of development; these concentrations are displayed in Table 3 (General Hydroponics, 2012).

Product	Amount Added
FloraGro	132 ml/100 liters
FloraMicro	264 ml/100 liters
FloraBloom	396 ml/100 liters

Table 3 Nutrients for Hydroponic System (General Hydroponics, 2012)

When transferring the plant from soil to hydroponic, it is necessary to remove as much soil as possible from the plant, because there are microbes in the soil can be harmful to the plants once in the hydroponic system. After removing the plants from the plastic planter, the soil mass is broken up. By doing this, the bulk amount of the soil is removed from the roots, as seen in Figure 12.





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Figure 12 Transferring Plant to Hydroponic System

After the bulk of the soil was removed, the root mass of the plant was placed in water to remove the rest of the soil. The plant was moved up and down in the water to remove the remaining soil mass is removed, as seen in Figure 13.





Figure 13 Transferring Plant to Hydroponic System

Prior to placing the plant into the CocoTek liner, it was half-filled with washed hydroton. The roots were spread out over the surface of hydroton. Hydroton was added to fill the CocoTek liner, seen in Figure 14.



Figure 14 Transferring Plant to Hydroponic System

After transferring the plant, the vortex sprayer was turned on. This process was repeated for each plant. Since there were only four plants in the hydroponic system, the remaining cups were filled with hydroton to prevent splashing of water outside the reservoir. The finished hydroponic system is displayed in Figure 15.



Figure 15 Finished Hydroponic System

RESULTS AND DISCUSSION

The uranium content in each sample was calculated using a comparative method described in the introduction. In each irradiation group, several check standards were irradiated as well; the results of which are displayed in Table 4. In the table, the expected results are displayed along with the observed results. For the uranium liquid standard, coal fly ash (NIST 1633B), and NORC, the expected and observed uranium concentrations agree well. However for the spinach leaves (NIST 1570), the uranium content within the spinach leaves was below detection limits. Values that were below detection limits were replaced with one-half the magnitude of the negative value (AlZahrani, 2010).

		U @ 106.1		U @ 277.6	
		ppm	±σ	ppm	±σ
U acetate	Stoichiometric	50.00		50.00	
U_STD-1	Observed	49.67	0.40	50.22	0.45
U_STD-3	Observed	50.47	0.42	50.81	0.48
NIST1633B	Consensus ¹	8.80	0.80	8.80	0.80
NIST1633B	Observed	7.76	0.17	7.70	0.33
NORC	Consensus ¹	2.96	0.34	2.96	0.34
NORC	Observed	2.56	0.08	2.90	0.18
NIST1570	Consensus ¹	0.46	3	0.46	3
NIST1570	Observed	0.225*	0.00	0.49*	0.01

Table 4 Results for Standards

¹ Glascock, M.D. (2006) Tables for Neutron Activation Analysis (6th edition), Research Reactor Center, University of Missouri-Columbia

* Negative replaced with one half the magnitude

Using the information from standard reference materials and Equation 4, uranium content for the soil samples was calculated and is displayed in Table 5. Results include the sample mass, the uranium content of the sample, and the standard deviation associated with the uranium content. It should be noted that for the hydroponic soil samples prior to contamination are soil samples not hydroton. Several

abbreviations are used in the table; they are the following: hydroponic control (CH), traditionally

contaminated control (CT), hydroponic (H), traditionally contaminated (TC), and one-time contaminated

(TO).

		106.1 keV		277.6 keV	
			Prior to Co	ontamination	
Sample	Mass (mg)	Uranium (ppm)	+/-σ	Uranium (ppm)	+/-σ
S-CH	248.7	0.7866377	0.0495488	1.0268660	0.1068533
S-CT	248.4	0.9272805	0.0444964	0.8132672	0.0954481
S-H1	251.5	0.6023712	0.0412406	0.7938980	0.0829245
S-H2	253.2	1.0057520	0.0521223	1.0749230	0.1029178
S-H3	252.0	0.7251220	0.0390455	0.6194296	0.0927481
S-H4	251.2	0.6842856	0.0435555	0.8273142	0.0908905
S-TC1	248.5	0.9244484	0.0546059	1.0228320	0.1081504
S-TC2	249.7	1.3840860	0.0518805	1.6039310	0.1123209
S-TC3	251.8	0.5609888	0.0424489	0.5762467	0.0859146
S-TC4	249.4	0.6343465	0.0495341	0.9261538	0.1054291
S-T01	249.8	1.0366410	0.0527714	1.0255640	0.1117135
S-T02	252.5	1.0956980	0.0512337	1.0735160	0.1043503
S-T03	248.8	0.6359463	0.0425187	0.6588511	0.0925423
		Post Contamination			
Sample		Uranium (ppm)	+/- σ	Uranium (ppm)	+/-σ
S-CH	252.0	0.97042140	0.0532891	1.02393700	0.10291470
S-CT	253.6	2.43777100	0.0840418	2.59397600	0.19171060
S-H1	250.4	2.44003300	0.1241903	2.58165800	0.22928180
S-H2	256.3	2.39962100	0.1028313	2.76142200	0.23105030
S-H3	254.2	2.12728900	0.1081866	2.66056400	0.18173590
S-H4	248.3	2.06957900	0.1263911	2.32386400	0.20433750
S-TC1	252.9	351.920900	3.370666	374.712600	6.375176
S-TC2	251.5	408.598300	3.909087	436.975300	7.427440
S-TC3	248.7	401.565600	3.841987	427.612900	7.269689
S-TC4	251.3	456.770600	4.366035	486.424300	8.262321
S-T01	250.9	854.653300	8.149809	894.831400	15.176550
S-T02	249.0	595.138500	5.681807	637.569800	10.821830
S-T03	251.5	567,102700	5,414402	605,411000	10,276210

Table 5 Results from Neutron Activation Analysis for Soil Samples

* Negative replaced with one half the magnitude

Using the information from standards and Equation 4, uranium content for the plant samples was

calculated and is displayed in Table 6. Results include the sample mass, the uranium content of the

sample, and the standard deviation associated with the uranium content. The same system of

abbreviations is utilized in this table.

		106.1 keV		277.6 keV	
	r	-	Prior to Co	ntamination	
Sample	Mass (mg)	Uranium (ppm)	+/-σ	Uranium (ppm)	+/-σ
P-CH	804.7	0.04453895*	5.617205E-04	0.04453895*	7.038739E-04
P-CT	796.6	0.18703560	0.02767053	0.20167890	0.03610151
P-H1	801.2	0.14031890	0.02436655	0.04590597*	7.254775E-04
P-H2	801.0	0.04453895*	5.966122E-04	0.04508805*	7.125516E-04
P-H3	797.2	0.04453895*	5.959052E-04	0.04683388*	7.401420E-04
P-H4	800.0	0.12630920	0.02658775	0.12260950	0.03131566
P-TC1	798.6	0.11834910	0.02591848	0.09891133	0.03044289
P-TC2	802.3	0.19892700	0.02936099	0.19860310	0.03559378
P-TC3	800.1	0.36022390	0.03275437	0.38331030	0.04104114
P-TC4	796.3	0.04453895*	5.982894E-04	0.04979246*	7.868980E-04
P-T01	800.1	0.18275810	0.02837830	0.30186730	0.03908770
P-T02	802.3	0.04453895*	6.226521E-04	0.04623432*	7.306667E-04
P-T03	798.6	0.04453895*	5.512424E-04	0.04496810*	7.106560E-04
		Post Contamination			
Sample		Uranium (ppm)	+/-σ	Uranium (ppm)	+/-σ
P-CH	801.2	0.2765132	0.0224651	0.2964715	0.0318651
P-CT	439.8	0.2402482	0.0353773	0.2041397	0.0511750
P-H1	800.0	0.5315588	0.0287913	0.6196845	0.0409024
P-H2	802.8	0.2436011	0.0271330	0.3376975	0.0390521
P-H3	697.5	0.2492374	0.0284030	0.3689731	0.0410748
P-H4	801.6	2.5282400	0.0415067	2.5273300	0.0524143
P-TC1	271.5	2.6087390	0.0632736	2.6424030	0.0815184
P-TC2	802.9	0.6061712	0.0295443	0.6319804	0.0392680
P-TC3	802.7	1.8994990	0.0330930	1.9883830	0.0481444
P-TC4	187.6	3.4652080	0.0719895	3.3996270	0.1037223
P-T01	522.5	1.0198660	0.0365151	1.0884790	0.0527558
P-T02	798.0	1.0084770	0.0295327	0.9967271	0.0385359
P-TO3	804.7	0.5492542	0.0237571	0.5878541	0.0307780

Table 6 Results from Neutron Activation Analysis for Pepper Samples

Prior to contamination, uranium content in soils averaged between 0.5 to 1.7 ppm (Table 5). This uranium content is consistent with average uranium concentrations in soil, which is around 2 ppm (International Atomic Energy Agency, 2012). The resulting pepper plant uptake was up to 0.4 ppm (Table

6). One thing to note is that the pepper plants were most likely grown for a longer period that the 34 day period of this study: starting as seed and progressing to mature pepper plants (APPENDIX A). This would have caused a larger uptake of uranium over the plant's cumulative life.

Concentration Ratios Using Soil Concentrations

The concentration ratio for pepper plants prior to contamination was calculated using Equation 1. The results of which are plotted on Figure 16 for the 106.1 keV and 277.6 keV peaks of Np-239. Negative values are the result of uranium content in peppers below detectability limits.



Figure 16 Concentration Ratio Before Contamination for Both Np-239 Peaks

For the 277.6 keV peak of Np-239, the pepper and soil uranium concentrations were overlaid on the concentration ratio data; this is displayed in Figure 17. In this case, uranium concentration in pepper plants is relatively constant, while soil content has more variance. For lower uranium concentrations in soil, this yields a higher concentration ratio, as seen in the case of plant TC3.



Figure 17 Concentration Ratio and Uranium Concentration for 277.6 keV Peak Before Contamination

After uranium contamination, uranium content in soils varied from 350 to 460 ppm for constant contamination, 560 to 860 ppm for one-time spike, and only 2 to 2.5 ppm for hydroponic systems (Table 5). While the pepper concentrations were more comparable, varying between 0.25 and 0.07 ppm (Table 6). The concentration ratio for pepper plants prior to contamination was calculated using Equation 1 and concentration ratios for Np-239's 106.1 and 277.6 keV peaks are displayed in Figure 18 and Figure 20. Figure 18 is an overview of all the contamination systems, while Figure 20 focuses on the traditionally contaminated systems. An overlay of uranium concentration information in soil and pepper samples is given in Figure 19 and Figure 21, which represent an overview and a focus on traditionally contaminated plants.



Figure 18 Concentration Ratio After Contamination for Both Np-239 Peaks

As seen by Figure 18, the concentration ratios are higher for hydroponic systems as well as controls. The reason for this trend is seen more easily by inspecting Figure 19. It should be noted that uranium concentration in this plot is a logarithmic scale, whereas Figure 17 has a linear scale. As seen by Figure 19, the uranium concentration in the pepper plants is relatively constant, but the uranium content in the growing medium increases significantly for soil samples versus hydroton samples. Thus although hydroponic systems had similar uranium concentrations in pepper fruit, they had larger concentration ratios due to the smaller concentrations in the hydroton of the hydroponic systems.



Figure 19 Concentration Ratio and Uranium Concentration for 277.6 keV Peak After Contamination

The traditionally contaminated plants had a concentration ratio around an order of magnitude less than the hydroponic systems. Between the groups, the constant contamination plants overall had a larger concentration ratio (Figure 20).



Figure 20 Concentration Ratio After Contamination for Both Np-239 Peaks - a More In-depth View

Between the constant contamination and one-time contamination group, the one-time contamination group had slightly higher uranium concentration in soil samples, as seen by Figure 21. This is interesting, because overall more uranium was applied to the constant contamination group. One possible explanation is that more uranium was absorbed into the pepper plant. This could be likely because overall peppers in the constant contamination group had higher uranium content.



Figure 21 Concentration Ratio and Uranium Concentration for 277.6 keV Peak After Contamination - a More In-depth View

Comparison of Concentration Ratios to Other Literature

Values for concentration ratios for the contaminated plants are listed in the following table. The hydroponic systems had the highest concentration ratios, between 1.12E-01 and 1.15E+00. While for the traditionally contaminated plants the lowest value for concentration ratios was 9.69E-04, and the highest was 7.59E-03.

Sample ID	106.1 keV Peak	277.6 keV Peak	Average	Group Average
H1	2.18E-01	2.40E-01	2.29E-01	
H2	1.02E-01	1.22E-01	1.12E-01	1 05F-01
H3	1.17E-01	1.39E-01	1.28E-01	4.032-01
H4	1.22E+00	1.09E+00	1.15E+00	
TC1	7.41E-03	7.05E-03	7.23E-03	
TC2	1.48E-03	1.45E-03	1.46E-03	5 17E-02
тсз	4.73E-03	4.65E-03	4.69E-03	5.172-05
TC4	7.59E-03	6.99E-03	7.29E-03	
TO1	1.19E-03	1.22E-03	1.20E-03	
TO2	1.69E-03	1.56E-03	1.63E-03	1.27E-03
TO3	9.69E-04	9.71E-04	9.70E-04	

Table 7 Concentration Ratios for Traditionally Contaminated Plants

The range of concentration ratios reported in the IAEA publication *Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater Environments* (Table 1) was 1.5E-2 and 2.0E-1 (International Atomic Energy Agency, 2010). The concentration ratios for the hydroponic systems were at the upper range or above the concentration ratio range in the IAEA publication. This suggests that for hydroponic systems, concentration ratios may not be an accurate measure of uptake of uranium based on availability. The range of concentration ratios for traditionally contaminated plants was within the range of the IAEA publication, but below the average value of 1.5E-02 (Table 1) (International Atomic Energy Agency, 2010). The lower concentration ratio could be due to the stunted growth of some of the pepper plants due to aphid activity. Additionally, the study was limited to 34 days and at the time of harvesting, not all the pepper plants had become fully grown.

Concentration Ratios Using Water Concentrations

In calculating the concentration ratio using water concentrations, it was assumed that the concentration of the water for the hydroponic system didn't change with time. For the traditionally grown systems the uranium concentration was averaged over the total volume of water added to the plant, such that the growing conditions had the following overall concentrations of uranium:

Growing Condition	Uranium Water Concentration
Hydroponic	50 μg/mL
Constant Contamination	25 μg/mL
One-Time Contamination	21.21 μg/mL

Table 8 Uranium Water Concentrations for Different Growing Conditions

The concentration ratio was calculated using Equation 2, the pepper plant concentrations and the water concentrations. Concentration ratios prior to contamination could not be calculated, because there was no uranium present in those water samples. The resulting after contamination concentration ratios are displayed in the following figure. The concentration ratios for the traditionally watered plants are mostly larger than the concentration ratios for the hydroponic plants. This is due to the water concentrations for each growing condition being comparative.



Figure 22 Concentration Ratios after Contamination using Water Concentration

The following figure is the concentration ratios using the 277.6 keV peak with pepper and water concentrations overlaid. It should be noted that pepper concentrations are in ppm, while water concentrations are in µg/mL. As seen by the figure, the water concentrations are fairly constant, while

the pepper plant concentrations vary for each condition. For the most part, the pepper concentrations



are greater for the traditionally contaminated systems than for the hydroponic systems.

Figure 23 Concentration Ratios after Contamination with Concentrations Overlaid using Water Concentration

CONCLUSIONS AND FUTURE WORK

While uranium uptake into the pepper plant fruit was comparable for each condition, the uranium uptake into the hydroton for the hydroponic systems was considerably less, yielding a lower concentration ratio when using soil concentrations. This could be due to the larger amounts of drainage in the hydroponic system or due to the inherent properties of the hydroton. When the concentration ratio was calculated using the water uranium concentration, resulting concentration ratios were similar for all growing conditions, but overall higher for traditionally contaminated systems. This was due to overall lower uranium concentrations in water for traditionally contaminated systems.

This suggests that for hydroponic systems, the concentration ratio using soil concentrations may not be appropriate for comparison with traditional systems and it may be more appropriate to use water concentrations. In future work, it would be beneficial to start the plants from seed. This would cause all uranium in the pepper plant to have been from the growing medium, i.e. soil or hydroton. In this current work, it's possible that some of the uranium in the peppers grown in hydroton could be from uranium already in the pepper plant translocated into the fruit. Another benefit is if two growing cycles were used (one before contamination and after), then the two groups would be comparable. More work would also need to be done on other radionuclides and plants.

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APPENDICES

APPENDIX A



TC2

TC3





