AN ABSTRACT OF THE THESIS OF

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Abstract approved:

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Uranium is the most common radionuclide that found in the nature. It is a radionuclide with a very low specific activity. There is limited data on plan uptake of uranium. This research was to determine the plant uptake of uranium, calculate the concentration ratio, and estimate the dose from eating contaminated radishes under certain conditions. The concentration ratio was pretty steady despite increasing the uranium in the soil by factor of 200. The concentration ratio ranges from 0.027 to 0.15. This concentration ratio could be used to calculate the uranium concentration in plants if uranium concentration in soil is known and other parameters stay the same. Also in this research the doses estimated from eating contaminated root vegetables exceeded the DOE and NRC limit of 1 mSv to general public.

Examination of Uranium Uptake by Root Vegetables and consequences for Human Consumption

by
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I understand that my thesis will become part of the permanent collection of Oregon State University libraries. My signature below authorizes release of my thesis to any reader upon request.

Abdullah Bakheet AlZahrani, Author

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1. Introduction

Oil and gas production and processing operations generate different types of waste (unwanted by-product). The most common classifications of oil and gas industry waste are hazardous, non-hazardous and radioactive waste. Naturally Occurring Radioactive Materials (NORM) waste is challenging oil and gas industries since it is not well regulated. For instance, at Saudi Aramco, the waste should contain activity of double background to be considered NORM waste otherwise the waste will be treated as hazardous or non-hazardous waste dependent on the chemical characteristics. So if the waste contains radiation less than twice the background, the waste will be treated as hazardous or non-hazardous waste and no further precaution will be taken regarding the radioactive contents. Oil and gas companies treat sludge in land farms where the soil and the sludge will be mixed, allowing bacteria to consume the organic components. This practice is repeatedly used in the same land farm, which could increase radiation levels over time even if the initial waste was not significantly enhanced in radioactivity. These locations can be sold to private investors who will use them for farming. However, once the oil companies do not survey the locations before sale to make sure they do not contain high levels of radioactive materials. There is little information regarding plant uptake of uranium and extensive work on plant uptake is needed to fulfill the data gap (IAEA-TECDOC-1616).

This research will add valuable information regarding plant uptake of uranium and uranium concentration ratios. Radishes will be used in this research as an example of root vegetables. This research will determine the radiation risk from eating radishes that are grown in contaminated soil. Also in this research, uranium concentration ratios will be calculated and compared to uranium concentration ratio from other literature to determine if the uranium concentration ratio can be used as a tool to predict the uranium uptake by plants. Finally, the internal doses that could result from consuming food produced at former land farms will be estimated.

The first use of “Radioactivity” was in 1899 when Curie used it in one of her publications even though Henri Becquerel had discovered that uranium is radioactive in 1896. He was studying the properties of X-rays using naturally fluorescent

minerals. This took place a few months after Roentgen discovered the x-rays. Becquerel thought that uranium emitted x-rays, after exposing potassium uranyl sulfate to sunlight. He used photographic plates wrapped in black paper to see the effect of the x-rays on them. First his research was not accepted in Paris in February 1986. Becquerel continued to develop his photographic plates and found that the images on the photographic plates were strong enough that he did not need to use the sunlight to produce images. Becquerel continued his research and came to the conclusion that the radiation emitted from uranium is not x-rays because the new radiation could be bent in a magnetic field

1.1 Uranium Radioactivity

In late 1780's Martin Klaproth discovered a black powder while he was studying pitchblende (which was thought to be an iron and zinc ore). Klaproth found that there was a portion of the ore that did not behave the way iron and zinc should behave. He named the new black powder uranium in honor of the planet Uranus, which was discovered in 1781. In fact this black powder is one of uranium oxides. The first time pure uranium was prepared was in 1841 when the French chemist Eugène-Melchior Peligot produced pure uranium from uranous chloride (UCl_4). Uranium is the heaviest naturally occurring element with an atomic number of 92. Pure uranium is a silver heavy metal. Table1 shows the properties of pure uranium, lead and gold.

Table 1.1: Uranium Properties adapted from The Encyclopedia of Earth (Fris, 2007)

Properties	U	Pb	Au
Color	Silver	Gray	Golden yellow
Density (g/cm3)	19.05	11.342	19.32
Melting point (K)	1405.2	600.702	1337.63
Boiling point (K)	4203	2024	3081
Electron Configuration	$[Rn]7s^2 5f^3 6d^1$	$[Xe]4f^{14} 5d^{10} 6s^2 6p^2$	$[Xe]6s^1 4f^{14} 5d^{10}$
Abundance in Earth's Crust (mg/kg)	2.7	1.4×10^{-1}	$4.0x10^{-3}$
Abundance in Earth's Ocean (mg/L)	3.2×10^{-3}	3×10^{-5}	$4.0x10^{-6}$
Abundance in Human Body (microgram)	200^1	140,000	Zero

Uranium is present in most soil and rock formations. The most common uranium ores,

both forms of uranium dioxide UO_2; are uraninite and pitchblende. The three naturally occurring isotopes of uranium are ^{234}U, ^{235}U, and ^{238}U. They are all alpha emitters and their abundances and radioactive characteristics are listed in Table 1.2

Table 1.2: Isotopic Composition of Naturally Occurring Uranium

Naturally Occurring Uranium (NDC, year)			
Isotope	Percentage	Half-life (Y)	Alpha [MeV]
U-238	99.28	4.468E+9	4.270
U-235	0.711	703.8E+6	4.679
U-234	0.0055	2.457E+5	4.859

Adapted from Nuclear Data Center, Japan Atomic Energy Agency

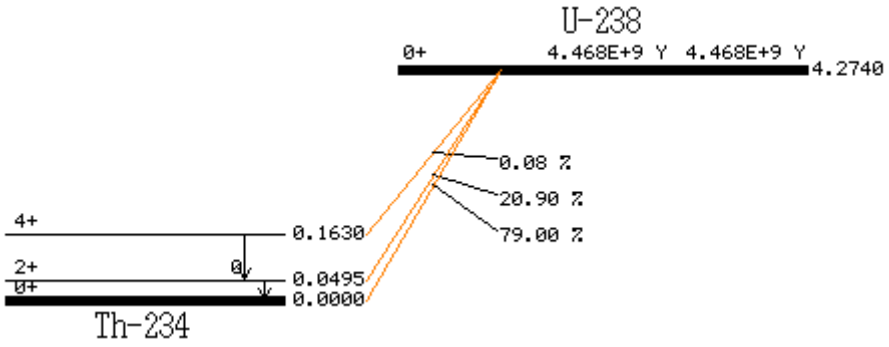
The most abundant isotope is ^{238}U. It decays through emission of alpha particles with 3 different kinetic energies and associated weak gamma rays as shown in Table 1.3.

Table 1.3: Alpha Emissions from ^{238}U

Mode of decay	Probability (%)	Alpha energy KeV	Gamma energy1 KeV	Gamma energy2 KeV
Alpha 0,0	77.54	4269.7	0.00	N/A
Alpha-0,1	22.33	4220.2	49.55	N/A
Alpha-0,2	0.13	4106.7	113.5	49.55

Adapted from Nuclear Data Center, Japan Atomic Energy Agency

238U A DECAY	Parent state: G.S.
Adapted from Table of Nuclides, Korean	Half life: 4.468E+9 Y(3)
Atomic Energy Agency;	Q(gs): 4274(5) keV
http://atom.kaeri.re.kr/index.html	Branch ratio: 1.0



Alpha ray:		**Gamma ray**:	
Energy(keV)	Intensity(rel)	Energy(keV)	Intensity(rel)
4198	79.0		
4151	20.9	49.55	*0.064*
4038	0.078	113.5	*0.0102*

Figure 1 Radioactive decay scheme of U-238

1.2 Health Effect of Exposure to Uranium

The potential adverse health effects associated with exposure to uranium are due to its radiological hazards and chemical toxicity. The radiological half-life of uranium is the period of time that the number of uranium atoms is reduced to a half whereas the biological half-life is a period of time that the number of uranium atoms entered the body is reduced to a half by discharging and decaying. The biological half life of uranium is shorter than its radiological half-life. Uranium biological half-life depends on the solubility of uranium compounds. Therefore, absorption decreases with decreasing solubility of the compound. Wrenn et al (1989) studied the uranium absorption in 12-volunteer adults by giving them uranium in drinking water; 40-50% of absorbed uranium from drinking water was excreted in the urine in three days after ingestion. The f_1 value for inorganic forms of uranium is about 0.02.

1.2.1 Radiological hazards

External exposure to radiation from pure uranium is less of a concern because the alpha particle emitted by its isotopes travel only a few centimeters in air or can be stopped by a sheet of paper. A relatively low concentration of U-235 (0.7%) in natural uranium emits a small amount of low-energy gamma radiation. However, internal alpha radiation exposure from uranium lodged in tissues is a more serious matter, since the adjacent tissues will be repeatedly irradiated.

Additionally, there is always also uranium progeny present. Within about a month, even purified uranium generates amounts of thorium-234 ($T_{1/2}$=24 d) and protactinium-234 ($T_{1/2}$=6.7 d) which emit beta particles (2.197 MeV of Pa-234) at almost the same rate as that of the alpha particles from the uranium-238. Two beta particles are emitted for each alpha particle. The radiological dangers of pure depleted uranium are lower (60 percent) than those of naturally-occurring uranium due to the removal of the more radioactive isotopes (U-235), as well as due to its long half-life (4.46 billion y).

The total annual intake of uranium is 460□g by ingestion and 0.6□g from by inhalation(Bem and Bou-Rabee, 2004). Recently it was reported only 22 microgram of uranium for people living in the USA and Europe (Friis, 2007); however, according to information available at the World Health Organization web page approximately 90 micrograms of natural uranium, on average; exist in the human body as a result of normal intake of water, food and air. The majority of this is found in the skeleton, with the rest in various organs and tissues.

1.2.2 Chemical Toxicity

The chemical toxicity of uranium is many times greater *in vivo* than its radiological hazard (Miller, 2002). Health effects of uranium are determined by factors such as the extent of exposure and whether it was internal or external. Two main pathways how an internal contamination of uranium may occur are inhalation and ingestion. Properties such as phase (e.g. particulate or gaseous), oxidation state (e.g.

metallic or ceramic), and the solubility of uranium and its compounds influence their absorption, distribution, translocation, elimination and the resulting toxicity. For example, metallic or insoluble form of uranium is relatively non-toxic compared to the soluble hexavalent uranium (VI) uranyl compounds (Gmelin, 1982).

Table 1.4 Selected Data on Uranium Toxicity [Craft et al, 2004]

Target area	Human studies	Animal studies
Renal	Elevated levels of protein excretion, urinary catalase and diuresis	Damage to Proximal convoluted tubules, necrotic cells cast from tubular epithelium, glomerular changes
Brain/Central Nerve System	Decreased performance on neurocognitive tests	Acute cholinergic toxicity; Dose-dependent accumulation in cortex, midbrain, and vermis; lectrophysiological changes in hippocampus
DNA	Increased reports of cancers	Increased urine mutagenicity and induction of tumors
Lungs/respiratory	No adverse health effects reported	Severe nasal congestion and hemorrage, lung lesions and fibrosis, edema and swelling, lung cancer
Gastrointestinal	Vomiting, diarrhea, albuminuria	n/a
Immune system	Chronic fatigue, rash, ear and eye infections, hair and weight loss, cough.	No studies
Reproductive	Uranium miners have more first born female children	Moderate to severe focal tubular atrophy; vacuolization of Leydig cells

2. Survey of the Literature

2.1 NORM in Oil and Gas Industry

Natural radiation exists everywhere. There are two sources of natural radiation, cosmic rays and naturally radioactive materials. The earth receives radiation from the sun and other stars within and beyond the galaxy; cosmic ray intensity increases as the

altitudes increases. The presence and abundance of naturally occurring radioactive material depends on the geology of the formation. The Earth's crust is radioactive and constantly releases radon gas into our atmosphere. The earth's soil contains uranium, thorium, and actinium in series. However, the neptunium series decayed away long ago because of the short half-life of neptunium ($T_{1/2}$= 2.14E6 years) relative to the age of the earth. There are also non series natural occurring isotopes such as potassium-40 ($T_{1/2}$=1.27E9), Cd-113 ($T_{1/2}$=9.3E15), and Gd-152 ($T_{1/2}$=1.08E14), (Martin, 2006).

The concentration of radioactive materials in soil varies from one location to another depending on soil type and the origin of the soil. Idaho State University calculated the activity of some nuclides in soil assuming that the density is 1.58 g/cm^3 and the volume is 7.894 E5 m^3

Table 2.1: Natural Radioactivity by the Square Mile, 1 Foot Deep

Nuclide	Activity used in calculation	Mass of Nuclide	Activity found in the volume of soil
Uranium	0.7 pCi/g (25 Bq/kg)	2,200 kg	0.8 curies (31 GBq)
Thorium	1.1 pCi/g (40 Bq/kg)	12,000 kg	1.4 curies (52 GBq)
Potassium 40	11 pCi/g (400 Bq/kg)	2000 kg	13 curies (500 GBq)
Radium	1.3 pCi/g (48 Bq/kg)	1.7 g	1.7 curies (63 GBq)
Radon	0.17 pCi/g (10 kBq/m^3) soil	11 µg	0.2 curies (7.4 GBq)
Total activity	>17 curies (>653 GBq)		

Human activities could elevate the levels of radiation above background levels. In such cases the material with elevated high radiation levels are called Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM).
In oil and gas industries, the NORM term is commonly used instead of TENORM because elevating the NORM levels is not intentional.

In oil and gas industries NORM can be enhanced during the process of producing natural gas and crude oil. The level of NORM could be several times higher than the background. However, in some cases the NORM levels can be elevated several hundred times the background level. Radionuclides of concern to the gas and oil

industry are uranium, radium-228, radium-226, radon-222, lead-210, and polonium-210 (Radiation Protection Manual, ARAMCO).

In oil and gas industries NORM can be deposited as scale in the pipes and equipment (Figure 2). Also it can be found in the sludge in tanks and vessels. According to the EPA, the petroleum industry generates about 260,000 metric tons of low-level radioactive waste (EPA, 2010). Radium-226, radium-228, lead-210, and polonium-210 are the radionuclides most commonly found in scale. Radium behaves chemically similar to calcium and barium forming radium sulfate scale. Radium-226 and radium-228 have been reported in scales as high as 405,000 pCi/g and 75,600 pCi/g respectively. Lead-210 and polonium-210 have very low specific activities in scale 2,025 pCi/g and 40.5 pCi/g. High concentrations of radionuclides have been reported in sludge and scrapings, the main radionuclides are Ra-226, Ra-228, Pb-210, and Po-210 (Radiation Protection Manual ARAMCO).

Table2.2: Range of activity concentration found in Scale, Sludge and pipeline scrapings

Radionuclide	Activity in Scale (pCi/g)		Activity in sludge (pCi/g)		Activity in pipeline scraping (pCi/g)	
	Min.	Max.	Min.	Max.	Min.	Max.
Ra-226	2.7	405000	1.35	21600	0.27	2025
Pb-210	0.54	2025	2.70	35100	1.35	1350
Po-210	0.54	40.5	0.11	4320	2.70	108
Ra-228	1.35	75600	13.50	1325	0.27	270

Adapted from Saudi Aramco Radiation Protection Manual

Figure 2: NORM Contaminated pipes (Department of Environmental Conservation)

Uranium has been found in the scraping of seawater pipelines in Saudi Aramco. Uranium (VI) is soluble in seawater ranging from 2 to 4 ppb (ppb=10E-9 g/g). According to Aramco uranium concentration was found to be about 5ppb in Arabian Gulf which is the major operation region for Aramco. Most oil companies consider the waste as NORM waste when radiation levels are at least double the background level. However, if the waste contains radiation less than the background, the waste will be treated as hazardous or non-hazardous waste and no further actions related to its radioactivity content will be taken.

2.2 Uptake of uranium by plants

Uranium is present in the soil and can be transferred from one environmental compartment to another. Plants may uptake uranium from the soil and humans may in turn consume the plants. According to the IAEA, the Soil-Plant-Man pathway is the major pathway for transfer of radionuclides to human beings. Uranium is an internal radiation hazard since it emits alpha, which cannot penetrate the layer of dead skin, and weak gamma rays as described in section 1.3. However, uranium is an internal radiation hazard when entering the human body due to the very high alpha energies emitted. The main pathway for uranium to enter human bodies is ingestion (drinking or eating contaminated water or food). EPA estimated that the daily intake of uranium from uncontaminated food ranges from 0.07 to 1.1 micrograms.

Uranium can be transferred through food chains and be consumed by human beings. The uranium transfer factor is the ratio of uranium concentration in plants to the uranium concentration in soil. The concentration ratios are widely used in radioecology to determine the plant radionuclide uptake. Meyer et. al (2004) gathered uranium CR data from various studies. He noted that the concentration ratio for grasses ranges from 0.025 to 0.110, for rice from 0.0019 to 0.44, for barley grain 0.0021 and 0.028 for barley stem. Also the CR is 0.00036 and 0.0048 for corn grain and corn stover, respectively. Another study by L.Laroche et al. (2005) showed that uranium concentrations in roots increased with increasing uranium concentration in

the solution. There was a linear relationship between uranium concentrations in the solution and total uranium in the roots. Uranium root uptake depends not only on free uranyl ions but depends on the total uranium in the solution. They noticed that uranium uptake was controlled by complexes at pH 7, hydroxocomplexes at pH 5.8, or by UO_2^{2+} at pH4.9. Finally, the presence of phosphate did not affect uranium uptake by Phaseolus vulgaris. Uranium solubility increased to more than 110 mg U/kg soil when citric acid was added to soil contaminated with uranium. Also citric acid increases plant uptake and the shoot uranium accumulation increases from 15 to 209 mg U/kg dried weight (Stephen et al., 1998). Also Pulhani et al (2005) studied uptake and distribution of naturally occurring uranium and other radionuclides in wheat plants from soil at two different locations in India. These locations contain uranium within the range of natural background (25 Bq/kg). Uranium concentration ranges from 9.4 to 42.2 Bq/kg. This study shows that the soil to wheat grain transfer factor is 4.0 E-4 to 2.1 E-3 for uranium-238. They also stated that the presence of calcium in soil increases the uptake of uranium. Moreover, high percentage of uranium is concentrated in the roots and very low percentage of uranium (1-2%) is in the grain. They concluded that the dose from consuming wheat grain from these two locations is very small and can be neglected. They also stated that the presence of calcium in soil increases the uptake of uranium. For instance, if the soil contains calcium, the ratio would increase as illustrated in equation 2.1

$$OR\ (U/Ca) = \frac{{}^{C_{Up}}/_{C_{Cap}}}{{}^{C_{Us}}/_{C_{Cas}}} \qquad \text{Equation 2.1}$$

where C_{UP} and C_{US} are concentrations of U in plant and soil, respectively, and C_{CaP} and C_{CaS} are concentrations of Ca in plant and soil, respectively.

3. Uranium Uptake in Radishes

3.1 Materials and methods

3.1.1 Soil preparation

Commercial grade potting mix from Corvallis Schmidt Garden Company was used in this study. It was prepared in 36 pots and weighed. Then the soil was allowed to dry out for 3 weeks in the main greenhouse at Oregon State University. The soil lost 48-49% of its initial weight. The soil data is shown in Table 3.1

Table 3.1: Soil weight

Sample	Wet Weight (g)	Dry Weight (g)	Sample	Wet weight (g)	Dry Weight (g)
1	216	114	C1	216	112
2	216	112	C2	218	112
3	218	116	C3	220	116
4	218	118	C4	216	118
5	216	112	C5	220	118
6	216	114	C6	224	120
A1	218	120	D1	222	118
A2	214	112	D2	224	116
A3	218	118	D3	222	118
A4	218	118	D4	216	114
A5	220	116	D5	220	114
A6	222	120	D6	222	118
B1	214	114	E1	224	122
B2	218	120	E2	220	118
B3	222	124	E3	218	116
B4	218	124	E4	220	120
B5	224	124	E5	222	116
B6	214	114	E6	218	116

3.1.2 Uranyl acetate

Uranyl acetate is a yellow rhombic crystal and it is not very soluble in water. The solubility is less than 10 g in 100 g of water. The molecular weight of uranyl acetate ($C_4H_{12}O_8U$) is 424.145 g/mol. The pH of 800 microgram/ml is 5.5 where it is 6.5 for 100 microgram/ml the other concentrations ranges between 6.5 to 7.

Uranyl acetate (UO2(CH3COO)2·2H2O) is a yellow free-flowing crystalline solid of yellow rhombic crystals .

This experiment was designed with 5 groups of radishes and a control group, each containing 6 replicate plants. The plants were watered with Uranyl acetate solutions of various concentrations. Each plant was watered three times a week with 100 ml of a previously prepared solution of the appropriate concentration.

Uranyl acetate was used in this experiment to prepare a stock solution containing 800 microgram/ml stock solutions of uranium. The total weight of uranyl acetate needed to prepare the stock solution in 500 ml flask was 713 mg. The stock solution was kept over 8 hours to allow uranyl acetate to completely dissolve. Then diluted solutions were prepared from the main stock solution as shown in the table below.

Table 3.2: Uranium concentration in solution & total uranium added for each group

Solution	U concentration (microgram/ml)	Total U added per 100 ml (microgram)
1	0 (Control Group)	0
2	1	100
3	5	500
4	10	1000
5	50	5000
6	200	20000

3.1.3 Radishes

A total of 6 groups containing 6 replicates of radishes were grown in the Radiation Center Greenhouse from seed purchased from an online see vendor.. Each

group was watered with a previously prepared uranyl acetate solution. Each plant was watered three times a week with 100 ml of the previously prepared solution of the appropriate concentration. The growing period was 4 weeks; each plant was watered 12 times with the proper solution.

Table 3.3 : Uranium concentration

Group	U Concentration (microgram/ml)	One time Total U (microgram)	One week Total U Microgram	One month Total U Microgram
Control	0	0	0	0
A	1	100	300	1200
B	5	500	1500	6000
C	10	1000	3000	12000
D	5	5000	15000	60000
E	200	20000	60000	240000

3.1.4 Harvest

The radishes were harvested after 30 days. The editable root portion of each radish was separated from the leaves and washed with distilled water. After that the roots were wiped off and weighed. Then all roots were put in the oven for more than 72 hours to dry at 55 C, as shown in the table 3.4

Table 3.4: Sample Masses or Radishes

Sample	Wet Weight (g)	Dry Weight (g)	Sample	Wet Weight (g)	Dry Weight (g)
1	6.789	0.477	C1	13.058	0.893
2	4.506	0.368	C2	9.935	0.844
3	7.687	0.606	C3	11.369	0.782
4	2.987	0.282	C4	13.197	0.91
5	7.031	0.593	C5	12.667	0.902
6	1.539	0.178	C6	14.613	0.974
A1	2.734	0.348	D1	6.095	0.517
A2	5.659	0.475	D2	10.353	0.75
A3	13.388	0.945	D3	14.609	0.919
A4	6.338	0.597	D4	4.134	0.309
A5	8.178	0.633	D5	0.748	0.062
A6	17.786	0.996	D6	3.182	0.377
B1	3.878	0.273	E1	14.814	0.932
B2	7.139	0.603	E2	2.347	0.277
B3	1.606	0.195	E3	2.424	0.329
B4	6.397	0.404	E4	7.399	0.665
B5	10.16	0.857	E5	6.048	0.476
B6	0.115	0.008	E6	5.968	0.501

4. Neutron and Neutron Activation Analysis

4.1 Introduction to Neutron

The neutron is the uncharged particle of the nucleon pair (neutron and proton), the only nucleus that does not have a neutron is ^{1}H. The first observation of the neutron was by Borthe and Becker when they struck beryllium target with alpha particles and obtained a very penetrating non-ionizing radiation. After that Curie and Joliot studied the new non-ionizing radiation and found that an energetic photon would be emitted if this type of radiation targeted paraffin. However, in 1932 Chadwick studied the new radiation and did additional recoil experiments and confirmed his hypothesis and discovered neutron as a non charged particle in the

nucleus. Neutrons disintegrate into protons and electrons in absence of other nuclear matters with half-life of 12.5 min but in fact materials will absorbed free neutrons before they decay away (Kryrger, 1971). Since neutrons have no charge we cannot accelerate them but we can only reduce their energy through collision with different materials.

Neutrons can be classified to four classes:

A) Thermal neutrons have energy about 0.025eV

B) Epithermal neutrons have energy about 1eV

C) Slow neutrons have energy about 1keV

D) Fast neutrons have energy above 100 kev

4.2 Neutron Sources

The only methods used to generate beams of neutrons are nuclear reactions. This section briefly discusses the main neutron sources:

Alpha- Beryllium Source

This is the first reaction used to produce neutron as mentioned above. In this reaction, the stable ^{9}Be will be mixed with alpha-emitting material such as ^{226}Ra to produce neutrons with varies energies up to 13 Mev. This reaction cannot be used to produce monoenergetic because the alpha-emitting material emits alphas with varies energies, alphas' energies can be reduced by collision with the materials, alphas will have different directions, and also C-14 might stay in an excited state, retaining some of the energy.

$$^{4}He + {}^{9}Be \rightarrow {}^{12}C + {}^{1}n$$

Photoneutron Source

Gamma rays can be used to eject neutrons from the target materials. In this process, using monoenergetic photons produces a beam of neutrons that will be close to monoenergetic.

$$\gamma + {}^{9}Be \rightarrow {}^{8}Be + {}^{1}n$$

Spontaneous Fission

Neutrons can be produced from the spontaneous fission of isotopes. Cf-252 is the most common isotope used to produce free neutrons. One gram of Cf-252 produces neutrons with rate of 2.3 E12 neutrons per second (Krane, 2008).

Nuclear reaction

Nuclear reactions can be used to eject neutrons from target materials. In this process, an accelerator should be used to produce particles that will eject neutrons from the target. If the particles are separated according to their energies and specific energetic particles are used, monoenergetic neutrons can be obtained.

$^{3}H + d \rightarrow {}^{4}He + n$

$^{4}He + {}^{9}Be \rightarrow {}^{12}C + n$

$^{7}Li + p \rightarrow {}^{7}Be + n$

Nuclear reactor

Nuclear reactors are also sources of neutrons; a typical reactor may have a flux of about 10E10 n/cm2/s, these neutrons can be moderated to thermal neutrons within the reactor. The fast neutron can be used to produce radioisotopes which is the application used in neutron activation.

4.3 Neutron Interaction

As a beam of neutrons travels, there are five reactions that can occur. Two of them are scattering reactions and the rest are absorption reactions.

A) Scattering reactions

Elastic Scattering

Elastic scattering occurs when a neutron strikes a nucleus and transfers some of its energy to the nucleus. This type of reaction most likely occurs when the neutron hits light nuclei. In this reaction no gamma rays will be emitted.

Inelastic Scattering

In inelastic scattering the neutron is not absorbed by the nucleus but transfers some of its energy to the nucleus. The product of inelastic scattering is a metastable isomer which will emit gamma rays and return to a stable state. This reaction happens when high-energy neutrons interact with heavy nuclei.

B) Absorption reactions

Neutron Capture

Neutron capture occurs when the neutron is absorbed by a nucleus and a gamma ray is emitted. Also beta- decay might occur . In these reactions, the mass number of the nucleus increases by one.

$$^{1}_{1}H+^{1}_{0}n \rightarrow [^{2}_{1}H] \rightarrow ^{2}_{1}H+\gamma$$

$$^{2}_{1}H+^{1}_{0}n \rightarrow [^{3}_{1}H] \rightarrow ^{3}_{1}H+\gamma$$

$$^{59}_{27}Co+^{1}_{0}n \rightarrow [^{60}_{27}Co] \rightarrow ^{60}_{27}Co+\gamma$$

Also beta- decay might occur after neutron capture

$$^{23}_{11}Na+^{1}_{0}n \rightarrow ^{24}_{11}Na+\gamma$$

$$^{24}_{11}Na \xrightarrow{\beta(1.39Mev)} ^{24}_{12}Mg^{*} \xrightarrow[\gamma(1.37MeV)]{\gamma(2.75MeV)} ^{24}_{12}Mg$$

Transmutation

When the neutron is absorbed by a nucleus the nucleus may emit charged particles. The most common emitted particles are protons, deuterons, and alpha particles.

$$^{1}_{0}n+^{16}_{8}O \rightarrow ^{16}_{7}N+^{1}_{1}P$$

$$^{1}_{0}n+^{10}_{5}B \rightarrow ^{7}_{3}Li+\alpha$$

Fission

Fission can occur when the neutron is absorbed by elements that have Z>90. The nucleus will split into large fragments with a release of two or three neutrons (Kryrger, 1971).

There are several analytical methods could be used to determine the uranium concentration in the samples. The most common analytical methods are ICP-MS and NAA. ICP-MS converts the atoms of the elements to ions, and then theses ions are separated and detected by the mass spectroscopy. The active film coating will be consumed when ions hit the surface of the detector so the dynode needs to be replaced within 6-18 months. Also, the detectors are sensitive to photons as they are to ions.

Moreover, NAA has the advantages of being a non-destructive technique and requiring little sample preparation whereas

4.4 Neutron Activation Analysis

In 1936 Hevesy and Levi discovered neutron activation when they found that rare earth elements became very radioactive after exposing them to neutron sources. After this discovery, they used nuclear reactions to sample and measure the induced radioactivity. Neutron Activation Analysis is better than chemical analytical methods for two main reasons; first NAA has higher sensitivities than chemical methods which can be detected as low as 10E-13 g. Second NAA doesn't destroy the samples which can be retained after the induced activity decays away (Lilley, 2007).

NAA requires a neutron source, gamma ray detectors, as well as understanding the reactions that occur in the reactor and the decay of activated elements. Activation time and decay time should be recorded. The mass of samples should also be recorded. When an atom is placed in a neutron flux, and absorbs a neutron, the atomic mass will increase by 1. The activated atom will emit one or more gamma rays with characteristic energies, which can be used to identify the irradiated atom. The activity A of the produced radioactive isotope can be calculated if the irradiation time t, neutron-capture cross section of the irradiated element (σ), decay constant of the produced isotope (λ), and neutron flux (φ) are known:

$$A = \lambda.N_2 = N_1 \sigma \varphi (1-^{e- \lambda t})$$

Where N_1 and N_2 are numbers of the irradiated atoms and number of the activated (radioactive) atoms, respectively.

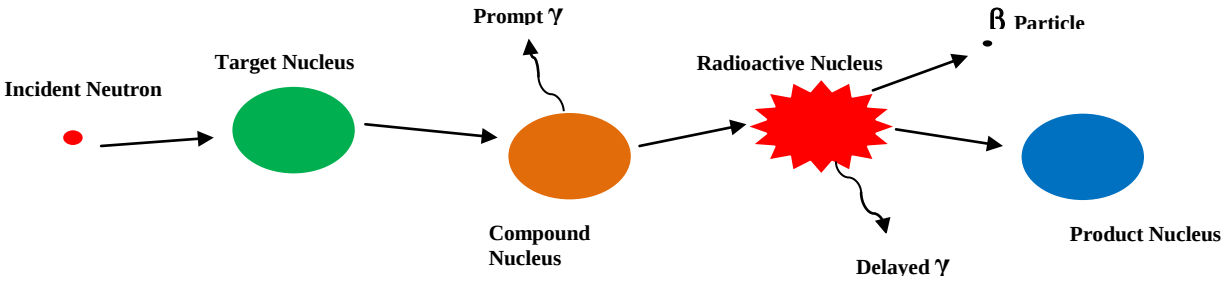


Figure 3: Neutron Activation Diagram

A nuclear reactor is the most common neutron source for neutron activation analysis. At Oregon State University, TRIGA Mark II, which is a pool-type reactor, uses uranium/zirconium hydride fuel elements in a circular grid array. The TRIGA can be operated at maximum steady state power of 1.1 MW and can be pulsed to 2500MW. U-235 is used to produce high energy neutrons ranging from 0 to 25 MeV. The emitted neutrons are ~~can be~~ moderated by water.

$$^{235}_{92}U + ^{1}_{0}n \rightarrow fission - prodcuts + 2.2n + energy$$

Then thermal neutron can be used to activate uranium-238

$$^{238}_{92}U + ^{1}_{0}n \rightarrow ^{239}_{92}U$$

$$^{239}_{92}U \rightarrow ^{239}_{93}Np$$

In this research the uranium concentration was calculated using standards containing a known uranium concentration. The known standard concentrations and the unknown sample concentrations were irradiated in the reactor. Then the activity of comparator standards and the unknown samples ~~will be~~ measured on the same detector. After that the equation below was used to calculate the mass of an element in the unknown sample relative to the comparator standards. Knowing the mass of the sample and the mass of the uranium can be used to calculate the specific activity of uranium in the samples.

$$C_s = C_{std} \frac{W_{std}}{W_s} \frac{A_s}{A_{std}}$$

A=activity of sample (s) and standard (*std*),

where c=concentration of uranium and W=weight of the sample and standard.

4.5 High Purity Germanium Detectors

Energy and gross counts are used to identify isotopes and quantify the amount of the isotopes. Radiation detectors should be energy sensitive to identify the isotopes and they should be able to count each event to quantify the amount of the isotopes.

High Purity Germanium (HPGe) detectors are one of the best radiation detectors; they use passive gamma ray emissions to identify isotopes. HPGe detectors have higher resolution compared to Sodium Iodide (NaI) detectors. NaI detectors perform poorly with mixed isotopes and high background radiation and so are not suitable for NAA.

5. Sample

5.1 Sample Processing

After drying each root was put in the mortar with liquid nitrogen added to freeze the root which makes it easy to crush the root. A pestle was used to ground the root. After grinding, the small particles were very well mixed. Then the roots were put in the oven for three days to assure that no moisture was absorbed while adding nitrogen liquid.

Leaves were also dried and crushed by hand then mixed well. The leaves were combined from each group to evaluate uranium uptake in leaves.

5.2 Samples and Neutron activation Analysis

About 100 mg of each sample was put in a vial and then it was sealed under a very high temperature. Also standards were prepared from the main stock solution (800microgram/ml) and other NIST1633A-1 and NIST1633A-2 standards were

prepared. The table below shows the weight of each sample. The samples were put in the reactor for neutron activation for 35 hours and then the samples were allowed to cool down. Three important energies were measured but in this research the gamma with 106.1 KeV was used to calculate uranium concentrations.

6. Results

The concentration ratio is pretty steady for the different groups even though the uranium concentrations in soil were increased by a factor of 200.

6.1 Uranium concentration in plant

Three high purity germanium detectors were used to detect gamma rays with three different energies. Two values were below the detection limit, these numbers were replaced by half the detection limit (0.04ppm) and designated (ND) in the table below (Antweiler and Tylor 2008; Flynn 2009; Helsel 2009).

Table 6.1: Root and Leaf Samples

Sample	**Mass (mg)**	**U (PPM)**		**Sample**	**Mass (mg)**	**U (PPM)**	
		106.1	**±*σ***			106.1Kev	**±*σ***
1	99.0	0.04 (ND)	0.00	C4	99.1	3.09	0.12
2	100.8	0.18	0.03	C5	101.9	5.58	0.21
3	114.7	0.26	0.03	C6	97.8	1.44	0.07
4	118.2	0.15	0.03	D1	100.0	9.45	0.34
5	102.7	0.04 (ND)	0.00	D2	102.8	19.03	0.68
6	99.7	0.37	0.04	D3	99.8	12.72	0.46
A1	103.4	0.63	0.04	D4	99.7	5.89	0.22
A2	103.2	0.30	0.03	D5	39.4	55.74	1.99
A3	101.9	0.39	0.03	D6	99.0	14.81	0.53
A4	102.2	0.43	0.03	E1	101.9	59.09	2.10
A5	102.7	0.94	0.05	E2	103.0	50.85	1.81
A6	100.3	0.39	0.04	E3	98.9	39.01	1.39
B1	101.3	2.56	0.10	E4	98.9	66.94	2.38
B2	99.3	1.49	0.07	E5	101.0	86.45	3.07
B3	101.4	2.46	0.10	E6	103.1	60.80	2.16
B4	101.7	2.93	0.12	Leaves	102.1	0.26	0.06
B5	99.1	0.52	0.05	Leaves A	101.1	0.93	0.07
B6	0.0	0.00	0.00	Leaves B	101.1	7.91	0.29
C1	101.1	1.64	0.08	Leaves C	98.4	3.87	0.16
C2	97.8	2.83	0.11	Leaves D	102.1	32.74	1.17
C3	102.2	4.34	0.16	Leaves E	102.6	46.40	1.64

6.2 Uranium concentration in soil

The uranium concentration in soil can be calculated from the volume of added uranium solution or can be detected using the NAA method. The table below shows the uranium concentrations using both approaches.

Table 6.2: Uranium Concentration in Soil

Sample ID	U mg/g in soil (Measured)	U mg/g in soil (Calculated)	Sample ID	U mg/g in soil (Measured)	U mg/g in soil (Calculated)
1	0.82	0.00	C1	106.58	107.14
2	0.88	0.00	C2	108.29	107.14
3	0.95	0.00	C3	165.65	103.45
4	0.95	0.00	C4	84.95	101.69
5	1.20	0.00	C5	66.63	101.69
6	2.04	0.00	C6	243.75	100.00
A1	18.52	10.00	D1	1250.31	508.47
A2	16.00	10.71	D2	278.18	517.24
A3	20.46	10.17	D3	131.11	508.47
A4	16.71	10.17	D4	911.12	526.32
A5	8.08	10.34	D5	232.95	526.32
A6	20.90	10.00	D6	904.51	508.47
B1	64.72	52.63	E1	1476.87	1967.21
B2	146.65	50.00	E2	233.77	2033.90
B3	62.48	48.39	E3	559.77	2068.97
B4	82.40	48.39	E4	640.60	2000.00
B5	57.18	48.39	E5	1050.31	2068.97
B6	114.04	52.63	E6	1906.01	2068.97

6.3 Uranium concentration Ratio

In this research, the calculated and measured uranium concentrations will be used to determine the ratio between uranium concentration in plants and uranium

concentration in soil. This assumes that total uranium will be in the plants or in the soil and no uranium will leach from the pots.

Table 6:3 Uranium Concentrations in Plant and Soil

Sample ID	U(μg/g) in soil	U μg/g in soil Cal.	μg/g in soil + Background	Sample ID	U(μg/g) in soil	U μg/g in soil Cal.	μγ/g in soil +Background
1	0.82	0	1.14	C1	106.58	107.14	108.28
2	0.88	0	1.14	C2	108.29	107.14	108.28
3	0.95	0	1.14	C3	165.65	103.45	104.59
4	0.95	0	1.14	C4	84.95	101.69	102.83
5	1.2	0	1.14	C5	66.63	101.69	102.83
6	2.04	0	1.14	C6	243.75	100	101.14
A1	18.52	10	11.14	D1	1250.31	508.47	509.61
A2	16	10.71	11.85	D2	278.18	517.24	518.38
A3	20.46	10.17	11.31	D3	131.11	508.47	509.61
A4	16.71	10.17	11.31	D4	911.12	526.32	527.46
A5	8.08	10.34	11.48	D5	232.95	526.32	527.46
A6	20.9	10	11.14	D6	904.51	508.47	509.61
B1	64.72	52.63	53.77	E1	1476.87	1967.21	1968.35
B2	146.65	50	51.14	E2	233.77	2033.9	2035.04
B3	62.48	48.39	49.53	E3	559.77	2068.97	2070.11
B4	82.4	48.39	49.53	E4	640.6	2000	2001.14
B5	57.18	48.39	49.53	E5	1050.31	2068.97	2070.11

Table 6.4: Average Uranium Concentration in Plant and Soil

Group	Average in Plant	Std Deviations	Average in Soil (Measured)	Std Deviations	Average in soil (Calculated)	Std Deviations
Control Group	0.173	0.128	1.14	0.460	1.14	0
Group A	0.513	0.236	16.8	4.69	11.4	0.267
Group B	1.99	0.980	82.7	37.0	50.7	1.85
Group C	3.15	1.59	129	65.2	105	3.01
Group D	19.6	18.3	618	462	517	8.77
Group E	60.5	16.0	978	626	2036	43.1

Table 6.5 : Uranium Concentration Ratio

Sample	Average Ratio Measured in plant + Measured in Soil	Average Ratio Measured in Plant + Calculated in Soil
Control Group	0.150	0.152
Group A	0.039	0.045
Group B	0.027	0.039
Group C	0.032	0.030
Group D	0.073	0.038
Group E	0.091	0.030

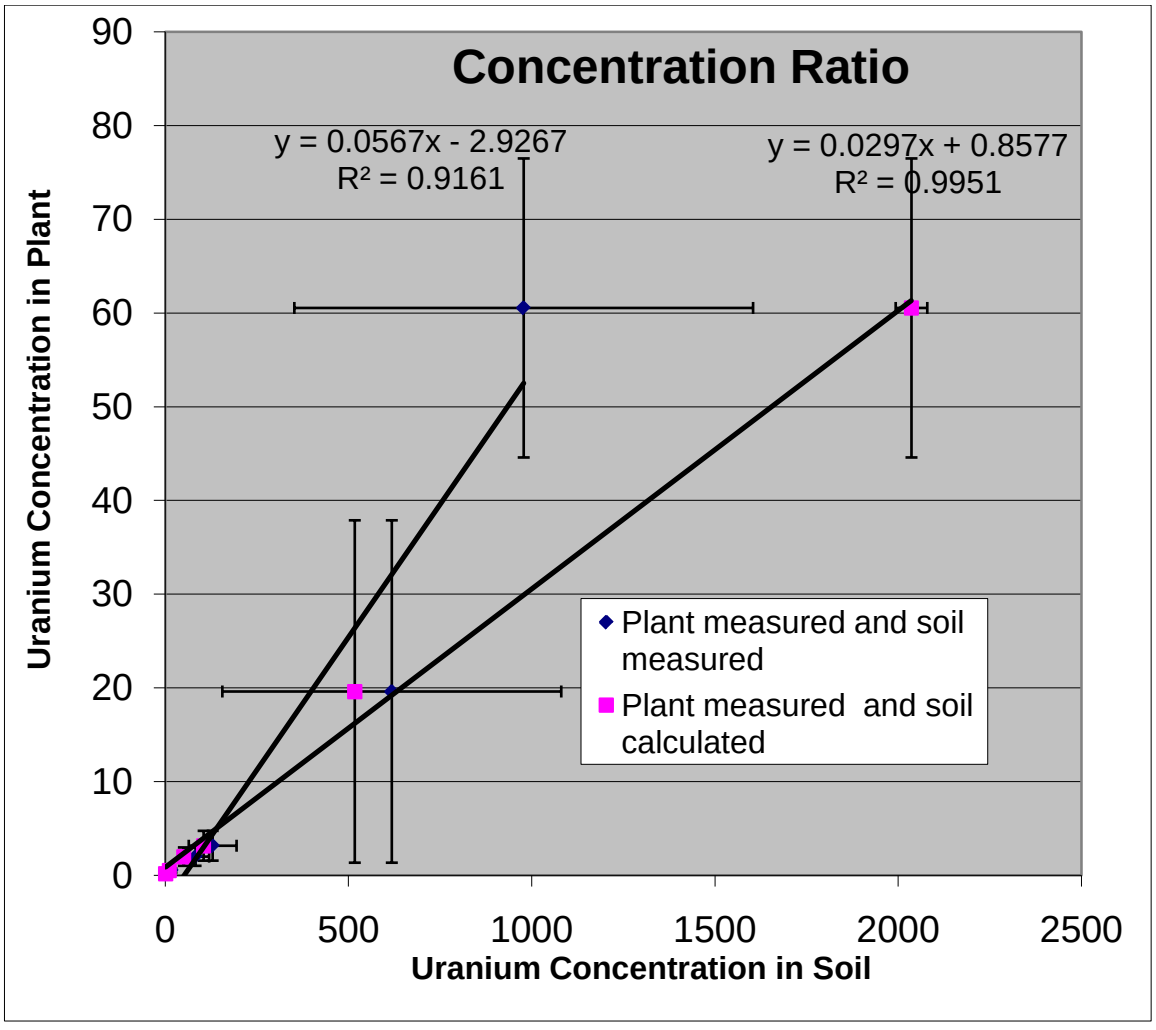


Chart 4: Average uranium concentration in plant and soil

7. Discussion

The uranium concentration ratio was pretty steady despite increasing the uranium concentration in soil by a factor of 200. Moreover, the CR ranges from 0.02 to 0.09 which is only a factor of 3. The study by Pulhani et al shows that the soil to wheat grain ranges from 4.0 E-4 to 2.1 E-3 which is a factor of 13.3. The CR could be used to estimate the uranium radish uptake for any given soil concentration if the soil and pH remain the same which could be a useful method to estimate the radiation dose from consuming radishes that are grown in contaminated soil. The average CR for group A is 0.04 +/- 0.04, group B is 0.03 +/- 0.02, group C is 0.03 +/- 0.03, group D is 0.07 +/- 0.09, and group E is 0.09 +/- 0.07. The standard deviation is high but this could be explained, being that healthy plants will consume more water and the uptake will be higher where less healthy plants would consume less water. For instance, plant B6 was unhealthy and the size of the plant after drying could not be weighted. This could be a biological plant behavior. The average CR for all the replicate radishes is 0.05 +/- 0.056 when using the measured uranium in soil, and it is 0.023 when using the calculated uranium in soil. The difference between uranium concentrations in soil using both approaches could be due to the uranium migration and leaching which will represent the actual environment scenarios. The concentration ratios might vary depending on many factors such as type of soil, type of plant, and essential nutrients. Also pH may slightly contribute to the differences in CR. For lower pH, the CR is three times higher than that for pH near 7.

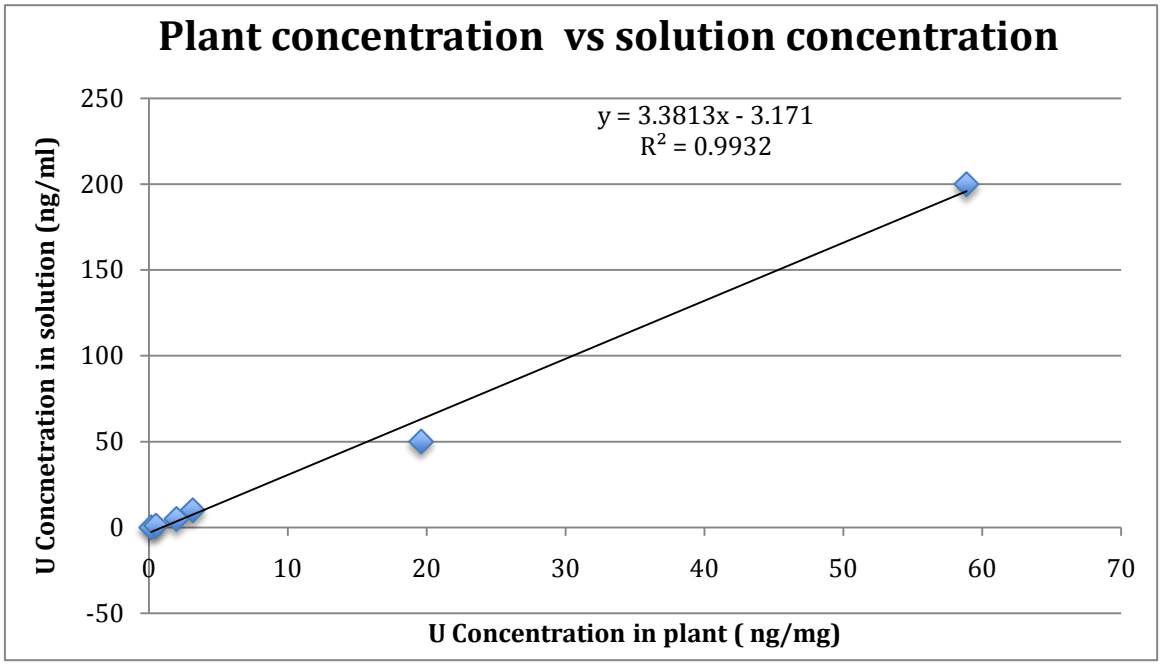


Chart 7.1: Uranium Concentration in Plant Vs Uranium Concentration in Solution

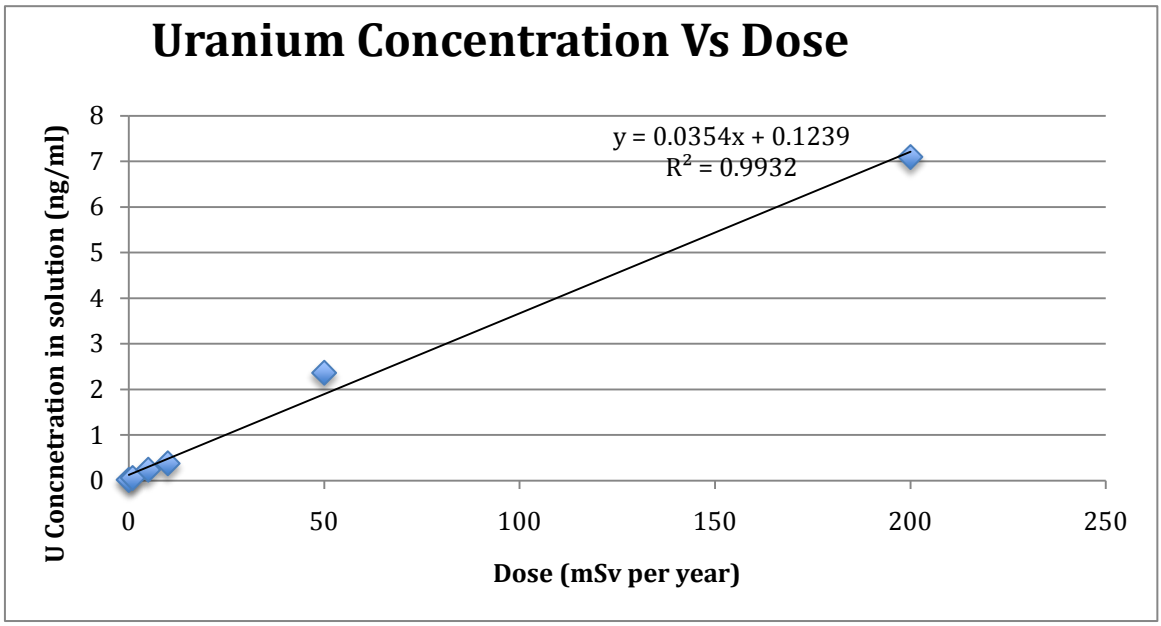
Dose estimation

This research is also designed to estimate radiation doses from eating uranium-contaminated food. The radiation doses from eating contaminated food can be estimated if the food consumption can be determined, and the dose conversion factor can be defined. The root vegetables consumption is 106 kg and the uranium conversion factor and specific activity are 4.5E-8 Sv/Bq ,and 25280 Bq/g, respectively. Table 6.1 shows the dose associated with the uranium concentration in plant and in the solution. This assumes that all root vegetables have the same concentration ratio and all root vegetables are grown in uranium contaminated soil.

Table 7.1: Estimated dose from consumption root vegetables

Group	Add uranium	Uranium in plant (ppm)	Dose (m Sv)	Added U (microgramg/ml)
Control Group	0	0.173	0.0209	0
Group A	1	0.513	0.0619	1
Group B	5	1.99	0.240	5
Group C	10	3.15	0.380	10
Group D	50	19.6	2.36	50
Group E	200	60.5	7.10	200

As shown on table 6.1, the Uranium concentration in plants increases with increasing the uranium concentration in soil in a direct variation relationship. The coefficient of determination is 0.993 which means 99.3% of the result can be explained by the linear line. Chart 6.1 shows how the data was fit in the linear relationship.



7.2 : Uranium Concentration Vs Dose

8. Conclusions and future work

The primarily goal of this thesis was to calculate the uranium concentration ratio and to estimate the dose from eating radishes that are grown in uranium contaminated soil. This research suggested that the uranium concentration ratio is pretty steady despite increasing the uranium concentration in soil by a factor of 200. Also, doses were estimated and it was estimated that people eating root vegetables grown in uranium-contaminated soil could receive doses higher than the limit of 1mSv.

This thesis focus on uranium uptake by radishes only, however, future work needs to be carried out for different radionuclide such as radium-226, radium-228, polonium-210, and lead 210. Also, the future work needs to consider different types of soil and how they might change the concentration ratios. Also, there is a general lack of available data in this area and different edible vegetables and fruits need to be studied to narrow this gap.

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