RADIUM AND RADON RADIATION RISK: AN EVALUATION OF THE YUCCA MOUNTAIN NUCLEAR WASTE REPOSITORY RISK ASSESSMENT USING DATA FROM NATURAL SYSTEMS

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Kelli Albertson

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The proposed high-level nuclear waste disposal site at Yucca Mountain, Nevada motivated a study of the source and transport of radium-226 through the geosphere and into the biosphere and the health effects related to radium-226 and its daughter product radon-222. The overall objective of this study was to grasp a better understanding of the long-term dose risk from radium-226 associated with the proposed high-level nuclear waste repository and to test the Department of Energy's Total System Performance Assessment (TSPA) risk modeling by using natural groundwater levels to simulate a potential release of radioactivity. Parameters and results of the TSPA were compared to natural groundwater concentration data for radium-226 from the United States Geological Survey National Water Quality Assessment data warehouse and from the literature. The

natural groundwater data containing radium permitted the comparison of calculated results to modeled data found in the TSPA.

A radioactive isotopic transport simulation was run using Mathematica software, showing a transport time for uranium-238 of 27,000 years, which is an order of magnitude faster than the result of 330,000 years obtained in the TSPA. Natural systems were analyzed and results showed that the systems were undersaturated with respect to both $RaSO_4$ and $RaCO_3$, two of the most common radium-compounds found in nature. Risk assessment calculations were performed using natural groundwater radium-226 concentrations, which were well below the United States Environmental Protection Agency (USEPA) maximum standard. Results suggest natural systems analyzed are well within the concentration limits allowed by the USEPA. A better understanding of the long-term dose risk from radium-226 associated with the proposed high-level nuclear waste repository was accomplished with this study.

CHAPTER I

INTRODUCTION

The proposed nuclear waste disposal site at Yucca Mountain, Nevada motivated a study, this thesis, of the release of radium-226 through the geosphere and into the biosphere as well as the health effects related to radium-226 and its daughter product radon-222. The Total System Performance Assessment (TSPA) produced by Sandia National Laboratory (SNL) for the Yucca Mountain Project (YMP) provided risk assessment data for the Department of Energy (DOE) repository license application to the Nuclear Regulatory Commission (NRC) (USDOE, 2008). This report shows that radium-226 becomes an important dose contributor after 200,000 years (Figure 1).

The overall objective of this study was to grasp a better understanding of the long-term dose risk from radium associated with the proposed high-level nuclear waste repository at Yucca Mountain, Nevada and to test the Yucca Mountain risk modeling by using natural groundwater levels to simulate a potential release of radioactivity. Parameters and results of the TSPA were compared to natural groundwater concentration data for radium from the United States Geological Survey (USGS) National Water Quality Assessment (NAWQA) data warehouse and from the literature (Almeida *et al*., 2004). The natural groundwater data containing radium permits comparison of natural data to predicted or modeled data found in the TSPA. Chapter II provides an overall background on radiation, nuclear waste, and the YMP. Chapter III (section 3.1) explains

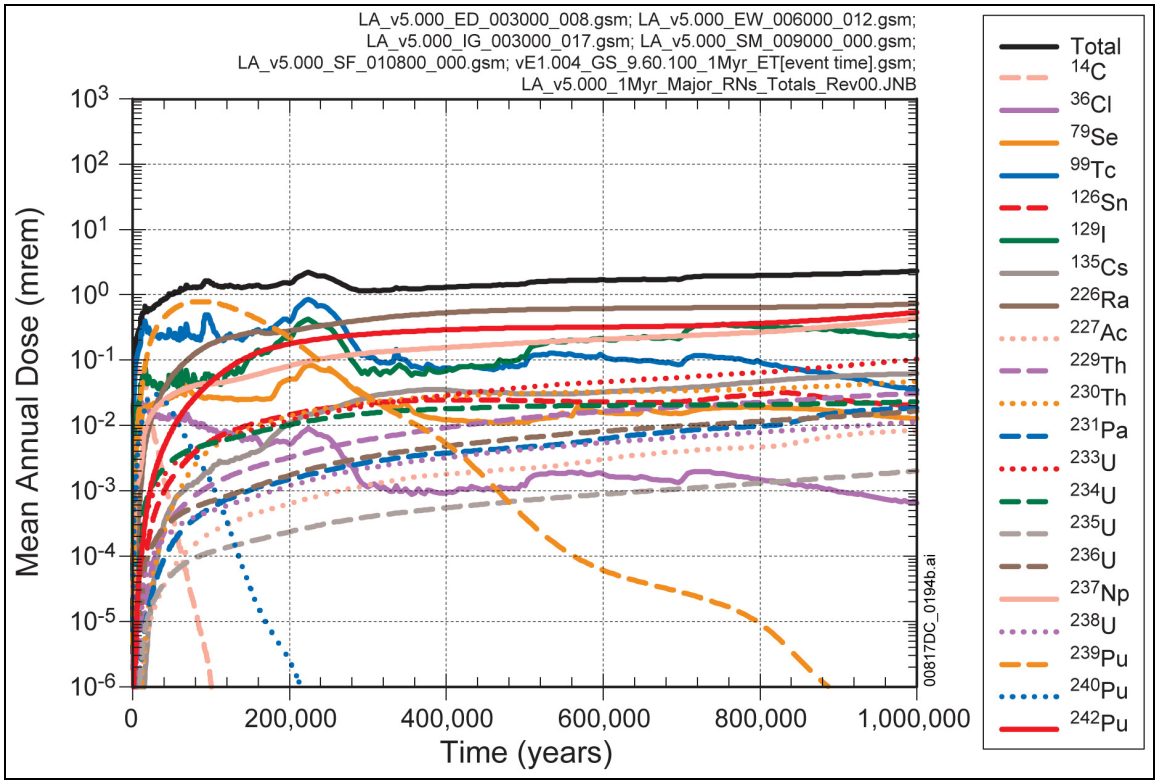

Figure 1. Total mean annual dose at 1,000,000 years: contribution from individual radionuclides (USDOE, 2008, V1pg1127).

radioactive isotopic transport and a transport simulation (Chapter IV, section 4.1) was run using Mathematica software (Wolfram Research, Inc., 2008). In Chapter IV (section 4.2), solubility limits were calculated for $RaSO_4$ and $RaCO_3$, two of the most common radium-compounds found in nature. Risk assessment calculations were performed and results are reported in Chapter IV (section 4.3). A conclusion and discussion are provided in Chapter V.

Nuclear waste is a worldwide issue. If not disposed of correctly it can be extremely harmful to humans. Health effects are the driving concern with respect to nuclear energy development and proliferation. The DOE, the United States Environmental Protection Agency (USEPA), and the NRC have set regulations and

standards for radioactive waste disposal, human dose, and environmental protection. The proposed nuclear waste repository at Yucca Mountain was set to house many radioactive isotopes (USDOE, 2008). In order to fully understand the release, transport, and fate of these isotopes many factors must be examined. They include a complete knowledge of the background of the YMP (Chapter II, section 2.2), a comprehensive understanding of the chemistry of radioactive isotopes (Chapter II, section 2.1.1), the mechanisms of release and transport of these isotopes through the geosphere and into the biosphere (Chapter III, section 3.1), parameters that effect transport (Chapter III, section 3.2), and their effects on human health and the environment once they enter the biosphere (Chapter III, section 3.3).

The TSPA results are derived from a complex and extensive statistical approach (Monte Carlo Method) with many uncertainties taking into account. This thesis used an analytical model; that was a relatively realistic simplified transport model with the most important processes accounted for (Chapter III, section 3.1).

CHAPTER II

LITERATURE REVIEW

2.1 Ionizing Radiation Background

Chapter II gives a brief history of the proposed nuclear waste disposal site at Yucca Mountain as well as general background knowledge of radioactive isotopes, radiation, and radiation risk. Nuclear waste issues surrounding Yucca Mountain are discussed and a political timeline is provided. Nuclear waste classifications, described in this chapter, give readers a better understanding of the types of nuclear waste currently being produced in the United States as well as the types of waste the proposed repository at Yucca Mountain was designed to house. A detailed description of the TSPA is given along with a discussion of the source, an idealized waste package (WP), and the inventory of radium at the proposed repository. A detailed explanation of the uranium decay series and the composition of the waste forms are also found in this chapter.

Nuclear waste is any solid, liquid, or gaseous material of no economic value or further use that contains radionuclides and emits energy, waves, or streams of energetic particles called radiation. Atoms are composed of a nucleus, stabilized by nuclear forces, carrying a positive charge and an outer electron shell that carries a negative charge. Naturally, some nuclei are stable, while others are unstable and decay forming new nuclei. When unstable nuclei decay, energy in the form of particles or waves is emitted as ionizing or non-ionizing radiation. Non-ionizing radiation is radiation

that lacks the energy to remove the outer electrons from atoms but can cause electrons to jump to higher energy states. Examples of non-ionizing radiation are: visible light, ultra-violet light, infrared radiation, microwaves, and radio waves. Biological effects, e.g. sunburns, are observed when there is too much exposure to non-ionizing radiation. Typically skin and eyes are most affected by non-ionizing radiation. In contrast, ionizing radiation has enough energy to remove electrons from an atom leaving charged particles (ions) behind. These charged ions are harmful to organisms and people, as they react chemically with other molecules and can damage living tissue and DNA (NRC, 2001a). In large amounts ionizing radiation can cause death, while in smaller amounts it can cause many types of cancer, tumors and genetic mutations (Hall, 2000). Humans are constantly exposed to ionizing radiation in small doses from the sun, watching TV, working on a computer, and other everyday activities. According to the World Health Organization, most human exposure to ionizing radiation comes from radon, the daughter product of radium (WHO, 2009).

There are three types of ionizing radiation that must be discussed when dealing with radioactive isotopes (Table 1). During the decay process, unstable isotopes will either decay through beta decay or alpha decay, giving off either beta radiation or alpha radiation. Each decay step also emits gamma radiation (NRC, 2001a). Different amounts of energy are associated with each type of ionizing radiation. An alpha particle is a form of radiation consisting of an energetic helium nucleus containing two protons and two neutrons (Figure 2). During an alpha decay, the atomic number of the parent decreases by two. Alpha radiation carries the least amount of energy and cannot penetrate paper (Figure 2). Beta decay is the release of an energetic electron and carries more

TABLE 1. TYPES OF IONIZING RADIATION

Type of Radiation	Physical Description
Alpha Radiation	An energetic particle that contains 2 protons and 2 neutrons (helium nucleus)
Beta Radiation	An electron
Gamma Radiation	An electromagnetic wave produced by sub-atomic particle interaction

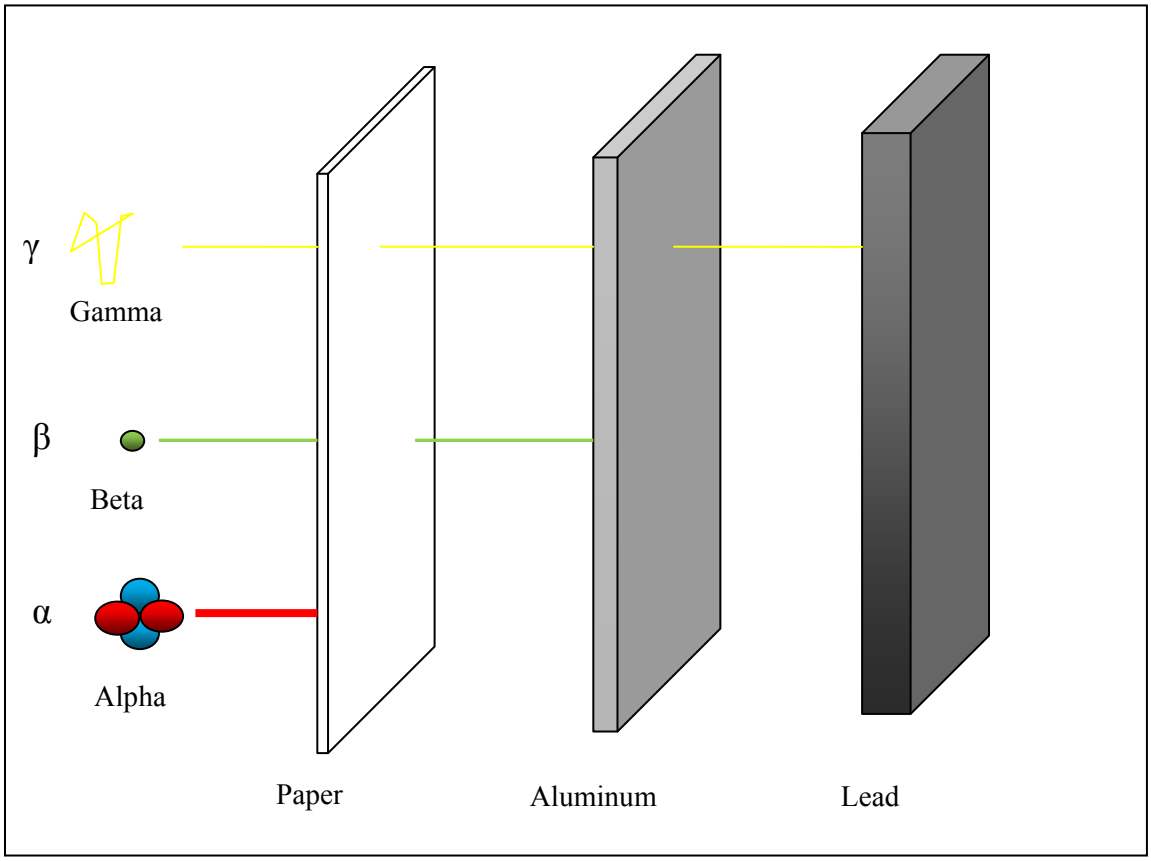

Figure 2. Representation of the relative strength of energy associated with each form of ionizing radiation.

energy than alpha radiation but will not penetrate aluminum (Figure 2). Gamma radiation carries the most energy and can penetrate materials thinner than lead (Figure 2).

2.1.1 Uranium Decay Series

This thesis examines the potential release of ionizing radiation from the proposed repository at Yucca Mountain. Uranium-238 is the main constituent in the nuclear waste that would be stored in the repository. The uranium-238 decay series goes through eight alpha decays and six beta decays and releases gamma radiation during all steps before ending with lead-206, a stable nucleus/nuclide (Figure 3). The two key isotopes in the uranium-238 decay series focused on in this thesis are radium-226 and radon-222 (Figure 3). Uranium-238 is an unstable isotope with a half-life of $4.47E^{09}$ years. It decays to thorium-234 releasing an alpha particle. Half-life describes the amount of time it takes for half of the original unstable nuclei to decay into new nuclei. From thorium-234, the series moves through two rapid beta decays to protactinium-234 and then to uranium-234. The decay of uranium-234, with a half-life of $2.46E^{05}$ years, releases an alpha particle resulting in thorium-230. Another alpha particle is released when thorium-230 decays to radium-226. Radium-226 then decays to radon-222 emitting an alpha particle. Radium-226 has a half-life of $1.599E^{03}$ years, while the half-life of radon-222 is only 3.8 days (USDOE, 2008).

2.2 Yucca Mountain Project

The Yucca Mountain Project has a long history that spans more than two decades. Nuclear waste has an even longer history of more than half a century. For decades the United States government has known high-level nuclear waste (HLW), discussed in detail

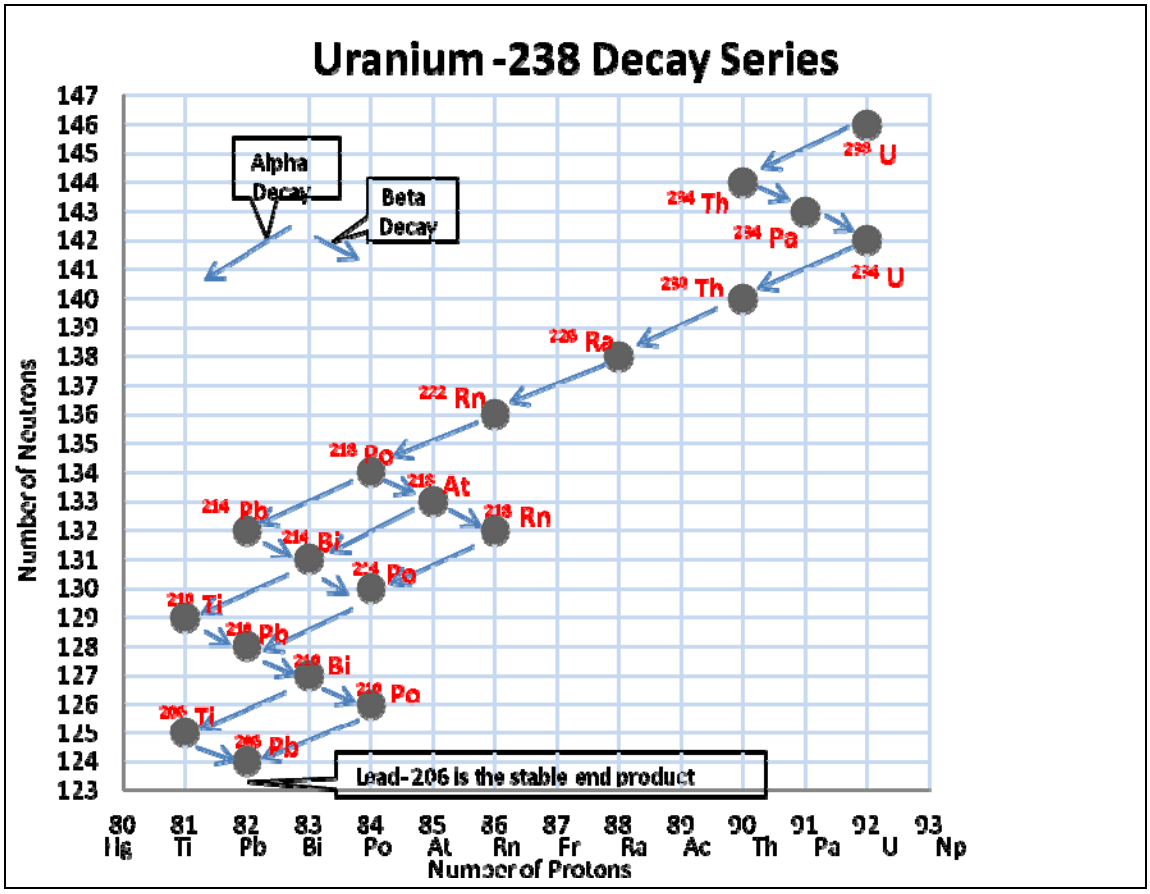

Figure 3. Nuclear decay series for uranium-238.

Numbers in red represent the mass number (i.e., neutrons plus protons). Arrows to the left represent alpha decays and arrows to the right represent beta decays. Down any decay path there are eight alpha decays and six beta decays. The series decays over time through numerous unstable isotopes until the series stops at lead-206, a stable isotope.

later in this chapter, would need a safe disposal site. The DOE first started looking at Yucca Mountain, Nevada as a proposed site for a HLW repository in 1978 along with eight other sites. In 1982, the DOE was given the responsibility of developing the repository in compliance with the Nuclear Waste Policy Act (NWPA). In 1987 Congress amended NWPA and choose Yucca Mountain as sole contender for the first geological repository for HLW in the United States. Decades later the site is still not open for the

disposal of waste. The next section discusses the political reasons for the YMP delays, the overall Acts, Policies and Regulations that affect nuclear waste, and a political timeline of major events since the DOE started looking for a disposal site. The political timeline is an updated and modified version of a table found in Uncertainty Underground by Macfarlane and Ewing (p. 47).

2.2.1 The Political Timeline (1974-2014)

- 1974- Energy Reorganization Act of 1974- established the NRC
- 1982- Nuclear Waste Policy Act- set roles for the DOE, USEPA, NRC
- 1983- NRC HLW regulations (10 CFR 60)
- 1984- DOE guidelines for site selection (10 CFR 960)
- 1985- USEPA Standards for HLW (40 CFR 191)
- 1987- Nuclear Waste Policy Act Amendments
- 1992- Energy Policy Act of 1992
- 1995- National Academy of Sciences- Technical Bases for Yucca Mountain Standards
- 2001- USEPA Standards for Yucca Mountain (40 CFR 197)
- 2001- NRC regulation for Yucca Mountain (10 CFR 63)
- 2001- DOE Yucca Mountain suitability criteria (10 CFR 963)
- 2002- President Bush notifies Congress of DOE selection of Yucca Mountain
- 2005- USEPA draft revised Standards (40 CFR 197)
- 2008, June- Submission to the NRC of the DOE license application for the repository construction authorization

- 2008, October- Final ruling from the USEPA on radiation standards for Yucca Mountain
- 2009- Obama Administration declaration that Yucca Mountain is "not an option"
- 2009, March- NCRP released report No. 160: updated average background radiation level
- 2010, March- Obama Administration eliminated funding related to the licensing proceeding for YMP
- 2010, June- DOE submitted a request to withdraw the application for the repository, motion denied, DOE appealed decision
- 2014- YMP is currently waiting to be addressed by the U.S. Court of Appeals for the District of Columbia Circuit.

The Energy Reorganization Act of 1974 established the Nuclear Regulatory Commission (NRC). The Act stated the commission would be composed of five members appointed by the President. The Act took all licensing and related regulatory functions from the Atomic Energy Commission and gave it to the NRC. The NRC would have the licensing and regulatory authority over the facilities used for storage of HLW, nuclear reactors (power plants), and any facility that is under contract with the DOE that fabricates nuclear reactor fuel. The NRC would be in charge of monitoring, testing, and recommending upgrades on all safety and safeguards of all facilities under its supervision.

The Nuclear Waste Policy Act (NWPA) was passed by Congress in 1982 and was amended in 1987. The NWPA specified roles agencies would have with regards to

different aspects of HLW. The DOE was given the role of dealing with aspects of the repository site: suitability, development, construction, and operation of the waste facility. The USEPA was put in charge of setting all health and environmental protection standards. The NRC was assigned the establishment of licensing requirements, granting the license itself, and implementing standards. The Act outlined guidelines for transportation of the waste, for choosing a repository site, and gave timeframes for dealing with radioactive waste.

In 1983, the NRC set general regulations for geological disposal of HLW (10 CFR 60). These standards established directives for all areas of the licensing processes from pre-application (site selection) through post-closure. The NRC set regulations for: design criteria of a geological repository, waste packages (WPs), violations of any regulations, participation from state governments, as well as the way records, reports, and inspections should be carried out. These regulations covered all aspects of a geological repository.

In 1984, the DOE set guidelines for selecting sites in the United States for HLW disposal (10 CFR 960), as required by NWPA. The guidelines for evaluating a site included: rock type, geohydrologic settings, geochemistry, environmental impacts of the site, population distribution and density around the proposed site, along with many more. These guidelines covered the entire site selection process.

In 1985, the USEPA set standards regarding management, storage, and disposal of HLW (40 CFR 191). However, the Natural Resources Defense Council (NRDC) brought suit against the USEPA stating these standards did not meet the Safe Drinking Water Act of 1974, insuring groundwater would not be endangered by

underground pollution. These standards were challenged, litigated, and reissued in December of 1993 (NRC, 1995).

In 1986, the DOE announced that from the nine original candidate geological repository sites they had narrowed it down to three; one in Washington, one in Texas and one in Nevada. Finally, in 1987 NWPA was amended putting Yucca Mountain, Nevada as the only site that would be pursued. The NWPA amendments established a Nuclear Waste Technical Review Board. The eleven-member Board, all appointed by the President, evaluates the scientific validity of activities regarding the site, WP's and the transportation of HLW. The Board reports to Congress with its findings, conclusions and recommendations and will be terminated one year after the repository starts to accept HLW.

The Energy Policy Act of 1992 (EnPA) stipulated a specific process for setting standards for the proposed repository. The Act required the USEPA to use analysis from the National Academy of Sciences (NAS) to set the environmental standards for Yucca Mountain. In 1993 the USEPA proposed set standards for the YMP, but had to vacate them due to a non-compliance with the recommendations of the NAS as required by this Act.

In 1995, NAS published its recommendations for the Technical Bases for Yucca Mountain Standards. Recommendations differed from the approach taken by the USEPA (40 CFR 191). They noted a 10,000 year time frame might not include the peak dose risk. NAS recommended a risk-based approach when dealing with adverse effects of human intrusion. NAS also stated: it is beyond the limits of scientific analysis to project the behavior of human society over long periods of time, assessments must be made with

substantial uncertainties, and scientifically justifiable analysis over many thousands of years could be performed with some degree of confidence. NAS recommended an acceptable level of risk, whether or not the development of the repository should proceed, and if Yucca Mountain would comply with the standards eventually adopted (NRC, 1995).

In 2001, the USEPA issued the Public Health and Environmental Radiation Protection Standards for Yucca Mountain, Nevada (40 CFR 197). This USEPA document established standards that required the DOE to limit radiation dose from the repository for 10,000 years. In 2004, portions of the 2001 standards, focusing on the time period for which compliance must be met, were removed by the US Court of Appeals D. C. circuit, *Nuclear Energy Institute, Inc. v. USEPA*. The 10,000 year compliance period was ruled not to be based on the recommendations by NAS and the court required that portions of the USEPA standards be reissued to a time frame that extended to 1,000,000 years.

In 2001, the NRC issued licensing standards for Yucca Mountain (10 CFR 63) (NRC, 2001b). The new standard (10 CFR 963) amended the original standard (10 CFR 960). That same year the DOE issued suitability criteria guidelines focused on the methodology that would be used for evaluating the geological repository (10 CFR 963).

In 2002, President George W. Bush notified Congress that he recommended Yucca Mountain, Nevada as a suitable site for the development and construction of the HLW repository. President Bush followed orders under the NWPA after he received the recommendation from the Secretary of Energy expressing confidence in the scientific and technical investigations that had occurred over the previous 20 years. Recommendation by the Secretary of Energy, the President's approval, and notification of Congress are

early steps in the process of selecting and completing a HLW repository mandated by the NWPA. The President's actions enabled the DOE to proceed with the federal licensing process required before beginning construction of the repository. The State of Nevada objected but Congress overruled it.

In 2005, in response to the 2004 ruling, the USEPA released new proposed radiation standards (40 CFR 197 revised) for Yucca Mountain. The USEPA set the annual exposure standard at 15 millirem (in addition to background radiation) for the first 10,000 years and at 350 millirem (in addition to background radiation) for 10,000 years up to 1,000,000 years. This new limit of 350 millirem, at the time, was almost equivalent to the average yearly exposure of an individual living in the United States of 360 millirem (USEPA, 2008).

In June of 2008, the DOE submitted a license application for the construction of the HLW repository to the NRC. The NRC was in charge of determining if the DOE had met all requirements and would decide if they would receive a license to construct the HLW repository.

The final rulings from the USEPA on dose standards for Yucca Mountain were announced in October of 2008. The USEPA did not change the original annual exposure standard set at 15 millirem (in addition to background radiation) for the first 10,000 years but did change the standard for 10,000 years up to 1,000,000 years to 100 millirem (in addition to background radiation) for the Reasonably Maximum Exposed Individual (RMEI). In response to the new standards, the Nevada Attorney General announced the state was filing suit to seek a court invalidation of the recently announced

USEPA radiation standard for the proposed HLW repository at Yucca Mountain (OHS, 2008).

In March 6, 2009, the Obama Administration declared Yucca Mountain as "not an option." This declaration stopped most of the work on the YMP.

In March 2009, the National Council on Radiation Protection & Measurements (NCRP) released report No. 160: Ionizing Radiation Exposure of the Population of the United States announced an updated average background radiation exposure for an individual living in the United States to 620 mrem.

In March 2010, President Obama announced the federal budget for the fiscal year beginning in October 2010. The new budget eliminated all funding related to the licensing proceeding for Yucca Mountain. After this announcement the DOE submitted a request to withdraw its application to construct the HLW repository at Yucca Mountain.

In June 2010, the panel of administrative judges for the NRC ruled that the DOE's motion to withdraw its application to construct a HLW repository at Yucca Mountain was denied. The DOE appealed the decision and it is currently in the U.S. Court of Appeals for the District of Columbia Circuit.

Currently the United States still has no HLW repository. As of October 2014, the YMP was held up in the court system. There are concerns about the safety of the repository. Over the past 20 years the proposed repository at Yucca Mountain has generated many studies and has raised many questions covering a wide range of geological, engineering and safety issues. The overall goal of these studies is to account for all processes that could contribute to radiation exposure from HLW. An overall

assessment (Total System Performance Assessment, TSPA) was performed by the DOE and is discussed below.

2.3 Total System Performance Assessment

The DOE is required to evaluate the post closure performance of Yucca Mountain to determine the potential human health impacts from potential radionuclide release. The NRC required the DOE to complete a performance assessment to estimate radiological exposures to the RMEI during the 1,000,000 year compliance period (USDOE, 2008). The Total System Performance Assessment (TSPA) model was developed to meet that requirement. The TSPA is a complex and comprehensive system analysis where models, which include all of the important parameters and processes, are used to predict the behavior of the system. These predicted behaviors can then be compared to specific performance standards (USDOE, 2007).

A Monte Carlo simulation-based method was used to create the TSPA. The Monte Carlo method used multiple calculations with random parameter values from within possible ranges for each of the processes modeled (USDOE, 2007). The TSPA was created using projected and predicted values for each model component, by integrating probabilistic calculations, the use of many assumptions, by defining parameters, and with analyses documented in other official projects (USDOE, 2008). The models were run to determine the overall probabilistic performance of the entire repository and to determine the radiological dose to the RMEI at a distance of 18 kilometers (the compliance boundary) south and down gradient of the repository, which

corresponds to the flow path of groundwater (USDOE, 2007, 2008; Macfarlene and Ewing, 2006).

The TSPA evaluated the repository at Yucca Mountain according to six major elements; Water flow on the surface and into the subsurface in the unsaturated zone (UZ); Disruptive events including natural events and human intrusion; The release of radionuclides from the engineered barrier system (EBS); The thermal and chemical environment in the EBS; The degradation of the engineered components containing waste (e.g., WPs); The potential radiation doses to an RMEI caused by the migration of radionuclides through the UZ, saturated zone (SZ), and into the biosphere (USDOE, 2007). The focus of this thesis is on the last element specifically looking at the potential uptake by humans of radium-226 and radon-222. The potential uptake by humans that could lead to radiation doses and possible health effects will be discussed in Chapter III (section 3.3). The TPSA is discussed in further detail in Chapter III (section 3.1).

2.4 Nuclear Waste Classifications

Radioactive waste originates from many sources ranging from mining of uranium, nuclear power generation, the medical industry and military industry, and from scientific research (NRC, 2001a). There are two forms of radioactive waste: Low-Level Waste (LLW) and High-Level Waste (HLW).

LLW can be defined as items contaminated with radioactive materials or items that have become radioactive from exposure to radiation. The materials can include protective clothing, rags, tools, mops, medical supplies, and laboratory test supplies. LLW is typically stored on-site at licensed facilities until it has decayed enough that it

can be disposed of in the general trash or it is shipped to a LLW disposal site (NRC, 2001a).

HLW includes highly radioactive materials produced as byproducts of nuclear reactions occurring in nuclear reactors (primarily spent nuclear fuel, SNF) and materials produced during the manufacturing of nuclear weapons (NRC, 2001a). HLW is currently stored at government and industrial sites until a permanent HLW disposal repository is built. The geological repository at Yucca Mountain is designed exclusively for HLW (USDOE, 2008). Radioactive waste producing functions are important to society. It is imperative that governments come to conclusions of how HLW will be disposed of. Currently in the United States most HLW is held in temporary facilities.

There are four categories of wastes for disposal at Yucca Mountain: commercial spent nuclear fuel (CSNF), DOE-owned HLW glass, DOE-owned spent nuclear fuel (DSNF), and naval SNF. All of this waste will be placed in one of two types of WPs: CSNF WPs or co-disposal (CDSP) WPs. The total number of WPs modeled in the TSPA was 11,629 (8,213 CSNF WPs and 3,416 CDSP WPs) (USDOE, 2008).

There are over 200 nuclides in SNF that could be disposed of in the proposed repository (USDOE, 2007). In the analysis performed in the TSPA 29 nuclides were chosen out of the 200 and labeled an “idealized waste package” (Table 2, USDOE, 2008). There are large amounts of uranium-238 present initially in the waste, compared to extremely small amounts of radium-226 (Table 3).

With such a large mass of uranium-238 (8.74E+07 grams) compared to the mass of radium-26 (2.11E-05 grams) per idealized WP, the ingrowth of radium-226 and its daughter radon-222 are potential hazards at long time scales. Ingrowth occurs as an

TABLE 2. IDEALIZED WASTE PACKAGE (WP) CONTENT IN GRAMS PER WP

Radionuclide	CSNF	DSNF	HLW
^{227}Ac	2.47E-06	1.22E-03	1.91E-04
^{241}Am	8.18E+03	2.18E+02	3.75E+01
^{243}Am	1.24E+03	6.73E+00	5.75E-01
^{14}C [a]	1.35E+00	1.81E+00	0.00E+00
^{36}Cl	3.23E+00	4.23E+00	0.00E+00
^{245}Cm	1.75E+01	9.25E-02	5.43E-02
^{135}Cs	4.36E+03	9.74E+01	1.27E+02
^{137}Cs	5.90E+03	9.72E+01	3.02E+02
^{129}I	1.73E+03	3.56E+01	7.27E+01
^{237}Np	4.57E+03	8.14E+01	9.95E+01
^{231}Pa	9.17E-03	2.14E+00	1.53E+00
^{238}Pu	1.52E+03	1.25E+01	3.91E+01
^{239}Pu	4.32E+04	2.21E+03	5.58E+02
^{240}Pu	2.05E+04	4.35E+02	4.61E+01
^{241}Pu	2.66E+03	2.92E+01	2.41E+03
^{242}Pu	5.28E+03	3.02E+01	3.89E+00
^{226}Ra	0.00E+00	4.57E-05	2.42E-05
^{228}Ra	0.00E+00	1.51E-05	6.00E-06
^{79}Se	4.19E+01	6.82E+00	7.01E+00
^{126}Sn	4.63E+02	9.40E+00	1.70E+01
^{90}Sr	2.49E+03	5.22E+01	1.74E+02
^{99}Tc	7.55E+03	1.58E+02	1.01E+03
^{229}Th	0.00E+00	3.24E-01	3.30E-03
^{230}Th	1.52E-01	1.18E-01	8.12E-04
^{232}Th	0.00E+00	2.17E+04	2.98E+04
^{232}U	1.02E-02	1.28E+00	4.08E-04
^{233}U	5.76E-02	5.38E+02	1.94E+01
^{234}U	1.75E+03	4.73E+02	2.33E+01
^{235}U	6.26E+04	2.51E+04	1.41E+03
^{236}U	3.84E+04	1.25E+03	5.99E+01
^{238}U	7.82E+06	6.84E+05	2.37E+05

Note: CSNF: commercial spent nuclear fuel. DSNF: DOE owned spent nuclear fuel. HLW: high-level waste. (SNL, 2007c; USDOE, 2008) [a]18 percent of ^{14}C for CSNF resides in the hardware outside of the cladding of the WP

TABLE 3. ^{226}Ra AND ^{238}U INVENTORY PER WASTE PACKAGE SHOWING THE AMOUNT OF DECAY AND INGROWTH EXPERIENCED IN THE FIRST 50 AND 87 YEARS OF STORAGE

Radionuclide	CSNF at 2067	CSNF after 50 Years	DSNF at 2030	DSNF after 87 Years	HLW at 2030	HLW after 87 Years
^{226}Ra	0.00E+00	1.29E-04	4.57E-05	1.80E-04	2.42E-05	2.68E-05
^{238}U	7.82E+06	7.82E+06	6.84E+05	6.84E+05	2.37E+05	2.37E+05

Note: CSNF: Commercial spent nuclear fuel- DSNF: DOE-owned spent nuclear fuel. (SNL, 2007c)

unstable nucleus decays into a daughter atom. Table 3 shows ingrowth of both radium-226 and uranium-238 over short periods of time after deposited into the repository. Concentrations of the daughter atoms increase while concentrations of the parent atoms decrease. This effect is not observable for uranium-238 over such a short time frame, as represented in Table 3, due to uranium-238's extremely large half-life. In some cases the daughter atoms are also unstable. Since ingrowth will occur over time, the concentrations of radioactive nuclides down the decay series of uranium-238 will increase (Figure 3). Ultimately the rate of ingrowth will be controlled by the concentration of the source uranium-238. When the daughter isotopes of a radionuclide have a half -life that is much shorter than the half -life of the parent isotopes, then the decay rate of the daughter becomes equal to the production rate of the daughter. This condition is known as secular equilibrium. In secular equilibrium, all decay rates for members of a decay series are equal. The concentrations of all daughter isotopes will increase with time until their activities are limited by the decay of uranium-238. In secular equilibrium all isotope activities are the same (Faure, 1998).

This thesis looks at a constant uranium-238 concentration source from the repository and does not look at individual WP failure. The constant concentration source

could come from one WP failure or multiple WP failures, the source and end concentration of uranium-238 are the only concerns in the evaluated transport equation in Chapter 4 (section 4.1). In general, the radionuclide inventory of each WP may be immediately available for transport upon WP failure. After WP failure, the waste form begins to degrade and can be released into the environment surrounding the repository. As the waste form degrades, waste colloids can form, and may facilitate the release of radionuclides from the breached WPs into the environment.

2.5 Radiation Health Risk

Once a WP fails, the health risk of exposure to radioactive materials is the major concern when dealing with disposal of radioactive waste. A basic definition of risk is the probability or chance of experiencing adverse consequences of a hazard. Hazard is a descriptive term used to describe a particular situation or substance that can cause harm (Slovic, 1987). Hazard is the "bad thing" (e.g., radiation, car crash, volcano) while risk is the probability and possible adverse effect from the "bad thing" (e.g., the probability or chance of cancer, death, destruction). Humans are subject to risks from hazards every day and could possibly come in contact with radium and radon via many ways (e.g., breathing air with high concentrations of radon, drinking groundwater that has either nuclide in it, eating food that has been grown with contaminated water, living on contaminated soil) (USDOE, 2007). The purpose of a geologic repository is to limit society's risks to radiation exposure. The evaluation of the possible risk of HLW radiation is done through a risk assessment (USEPA, 1991). Chapter III (section 3.3) will discuss in detail the risk

assessment done by the DOE on Yucca Mountain. The following sections will discuss the health effects of exposure to radium and radon.

The NRC estimates the average human living in the United States is exposed to about 620mrem of radiation per year (NCRP, 2009). A rem or Roentgen Equivalent Man (rem) is the standard unit of dose equivalent used in the United States. The System International (SI) unit for dose equivalent is known as the Sievert (Sv). One Sievert is equal to one hundred rems. Both a rem and an Sv measure the effective dose of radiation on living tissue. There are additional units used when discussing radiation (Table 4). Contributing factors affecting the exposure rate for an individual include: frequent air travel, mine working, elevation of residence, nuclear medicine and use of medical X-rays.

TABLE 4. UNITS OF RADIATION

Units	Unit Association
Curie (Ci) Becquerel (Bq)* = 1 decay / second	Units associated with the measurement of the activity (rate of radioactive decay) as a number of disintegrations per second in a particular sample of radioactive material. 1Ci= 37 billion Bq
Radiation absorbed dose (rad) Gray (Gy) *	Units associated with the measurement of the energy of the radiation absorbed by a target material 1Gy = 100 rad
Roentgen equivalent man (rem) Sievert (Sv)*	Units associated with the measurement of effective dose of radiation on living tissue. 1 Sv= 100 rem

Note: System International units (SI)

One concern for an individual, or in the case of the TSPA a reasonably maximum exposed individual (RMEI), exposed to radium-226 or radon-222 is that both isotopes are only about half way down the uranium-238 decay series (Figure 3). Once

these nuclides are in the body, the decay to daughter isotopes continues. Radium and radon are both alpha emitters, meaning when they decay the radiation emitted is in the form of alpha radiation (Table 5). The RMEI will continue to receive radiation from alpha, beta, and gamma radiation down the uranium-238 series. This radiation is generated inside their body and is a health hazard.

TABLE 5. BASIC PROPERTIES OF RADIUM AND RADON

Element Name	Atomic Number	Series	Isotope of Concern	Radioactive Half-life (yr)	Radiation Type	USEPA MCL	Carcinogenic Class
Radium	88	Alkali Earth Metal	Ra-226	1.60E+03	Alpha emitter	5pCi/L	A
Radon	86	Noble Gas	Rn-222	1.04E-02	Alpha emitter	4pCi/L	A

Note: MCL: maximum concentration level (USEPA, 2012)

2.5.1 Health Effects: Radon-222

According to the NRC over half of the radiation exposure of an average individual living in the United States comes from radon-222. Radon-222 is a leading cause in lung cancer worldwide (WHO, 2009). Radon is a noble gas found in the air, rocks, water, and food. Radon-222 is the daughter isotope of radium-226 in the uranium decay series (Figure 3). Both the USEPA and the International Agency for Research on Cancer classify radon as a human carcinogen (USEPA, 1986; IARC, 1988; see carcinogenic classifications later in this chapter). Head sinus carcinoma is attributed to radium-226 decay to radon-222. Since radon-222 is a gas, it diffuses into the sinus cavity causing the irradiation of the epithelial lining cells by the alpha particles emitted during

the radon-222 decay process (Rowland et al., 1978; Carnes et al., 1997; Harrison and Muirhead, 2003).

2.5.2 Health Effects: Radium-226

Radium is an alkaline earth element and is found all over the world in water, rocks, bones, and drywall (WHO, 2009). The radium ion in nature is found in its cation (II) state. Radium is commonly found interchanged in the mineral structures of minerals containing any of the alkaline earth elements (e.g., calcium, barium, magnesium, strontium, and beryllium).

Radium metabolizes like calcium in the body, and is predominantly retained in the skeleton. Radium is initially deposited on bone surfaces but over time it is eventually incorporated into the interior of the bone (Leggett et al., 1982). As radium-226 decays to radon-222 it emits alpha particles. The alpha particles have a short range (40-50 μm) in soft tissue and even a shorter range in bone. Due to this short range, bone cancer usually develops on or close to the surface of the bone, caused by the decay of radium near the endosteal surface (Harrison and Muirhead, 2003). According to Harrison and Muirhead (2003) both radium isotopes 226 and 228 have been found to cause bone tumors.

Two additional health issues associated with the ingestion of radium are osteonecrosis (bone loss) and anemia. Damaging mutations associated with the constant emission of alpha particles can cause Osteonecrosis. The incorporation of radium into the bone can interfere with the marrow and the body's ability to produce red and white blood cells. Anemia corresponds to the loss of red blood cells and immune-deficiencies are associated with decreased white blood cell counts. The loss of red and white blood cells

can be caused by radium consumption (Harrison and Muirhead, 2003), therefore radium exposure can reduce one's ability to fight off various types of infections. There is also evidence that drinking water contaminated with high levels of radium can lead to oral, esophagus, and pharynx cancer in males (Hirunwatthanakul et al., 2006).

There are well-documented epidemiological studies of the health effects of radium on women from the early 20th century. This group of women studied is known as the Radium Girls. The women were employed in watch factories where they hand painted watch dials with paint that contained radium salts. The radium paint gave off a light blue glow and was found useful on watch dials and gauges allowing for viewing in low light conditions. Female workers were constantly exposed radiation due to the close proximity of the paint with their skin. Even more important was their oral ingestion of small amounts of paint during the practice of lip pointing, a technique where the worker would pinch the paintbrush between theirs lips in order to create a fine tip. Almost all of the dial painters were women of childbearing age. Given the unique characteristics of the exposed population, much data has since been accumulated on the Radium Girls (Fry, 1998).

Within a few years the women began showing symptoms of bone sarcomas, particularly in the jaw and sinuses. One study, Rowland et al., 1983, found that being employed for five s in the dial painting industry led to a 100-fold increase in the likelihood of developing bone sarcomas when compared to the rest of the population. When modeling the toxicology of radium, the study concluded that there was a linear dose-response relationship for bone sarcomas and the Radium Girls (Rowland et al., 1983).

There are also studies of these women that provide non-cancer data for radium exposure. In one cohort study, data taken from surviving former dial painters, found that women subjected to higher dosages of radium exposure (over 5 Sv) showed decreased fertility levels. The measured fertility reduction was found to be statistically significant in both the number of pregnancies and live births the women produced, when compared to average American women at the time (Schieve et al., 1997).

In 1966, an epidemiological study was conducted in the Mid-West dealing with ingestion of groundwater where radium was present. In this case-control study towns in Iowa and Illinois were monitored for mortality data. The exposed populations lived in towns whose drinking water contained greater than 3 pCi/L of radium-226, and the controls were the nearby towns with less than 1 pCi/L. The study found no significant difference in mortality rates between the exposed/non-exposed populations, however the exposed populations did show significantly higher rates of deaths coded for bone cancer when compared to control groups (Petersen et al., 1966). It should be noted that the average radium levels in drinking water of the exposed populations was 4.7 pCi/L; the current USEPA groudwater drinking standard for radium is 5 pCi/L.

2.5.3 Carcinogenic Classifications

The guidelines for carcinogen risk assessment classify chemicals and other agents into five groups based on scientific evidence for carcinogenicity. The groups and definitions are listed below (USEPA, 1986):

- Class A- Human Carcinogen- there is enough evidence to conclude that it could cause cancer in humans.

- Class B- Probable Human Carcinogen- there is limited evidence to conclude that it causes cancer in humans but evidence is not conclusive.
- Class C- Possible Human Carcinogen- there is limited evidence that it causes cancer in animals, there is a lack of human data, but evidence is not conclusive.
- Class D- Not Classifiable as to Human Carcinogenicity- there is no evidence that it causes cancer in humans.
- Class E- Evidence of Non-Carcinogenicity for Humans- there is strong evidence that it does not cause cancer in humans.

Radium and radon are both listed by the USEPA as human carcinogens (Class A). Currently there is enough scientific evidence to prove that if exposed to these radioactive elements cancer could be an outcome from the exposure.

CHAPTER III

METHODS

3.1 Nuclear Waste Transport

This section describes the physical setting of the Yucca Mountain including the geology, hydrology, and geochemistry of the surrounding area. The unsaturated zone (UZ) and saturated zone (SZ) will be explained in context of how each affects the transport of radionuclides. A detailed description of the transport of radionuclides from the proposed repository will also be presented. Both the DOE transport model and the model used in this thesis will be explained.

The release of radionuclides into the geosphere will occur after the WPs fail. Upon failure, nuclides will be transported down hydraulic and chemical potential gradients by advection and diffusion. The nuclides will eventually enter the SZ, below Yucca Mountain, and will be transported with groundwater flow down gradient. Mechanisms that will control the transport include source concentrations (including radionuclide ingrowth and decay), aqueous speciation, groundwater flow, colloid capture, matrix-fracture interactions, geochemistry of host soil/strata, and sorption and/or precipitation. Each of these mechanisms will play a role in the concentrations of aqueous radium-226 and radon-222 as they travel into the biosphere. In order to fully understand the mechanisms of release and transport a basic understanding of the environment surrounding the repository is needed.

3.1.1 Geology

The proposed repository is located on federal land near the military nuclear test site in Nye County, Nevada (36° 51'10" N, 116° 25'36" W). It sits about 100 miles northwest of the metropolitan area of Las Vegas, near the border of California and Nevada.

The modern-day climate of Nevada is semiarid to arid, with low precipitation and dry winds. There is a rain shadow effect caused by the mountains to the west of Yucca Mountain. The TSPA identifies the climate factors that will affect the water-transport processes in the UZ around Yucca Mountain as: solar radiation, diurnal and seasonal temperature cycles, relative humidity, precipitation, and extended periods of drought (USDOE, 2008).

The topography in the region around Yucca Mountain is characterized by extensive valleys and long and narrow, approximately north-south-trending, mountain ranges. Yucca Mountain is located in a transition zone between the Mojave Desert and the Great Basin Desert indicated by the red circle on Figure 4. Erosional processes have shaped the eastward facing slope of Yucca Mountain (USDOE, 2008).

Yucca Mountain consists of an uplifted, block-faulted ridge with alternating layers of volcanic welded and nonwelded tuffs of Miocene age (USDOE, 2008). There are three major geologic units in Yucca Mountain: the volcanic tuff formations of the Crater Flat Group, Paintbrush Group, and the Calico Hills Formation (Figure 5). There are four major hydrogeologic units: the Tiva Canyon welded, the Paintbrush non-welded, the Topopah Spring welded, and the Crater Flat undifferentiated units (Ortiz et al., 1985).

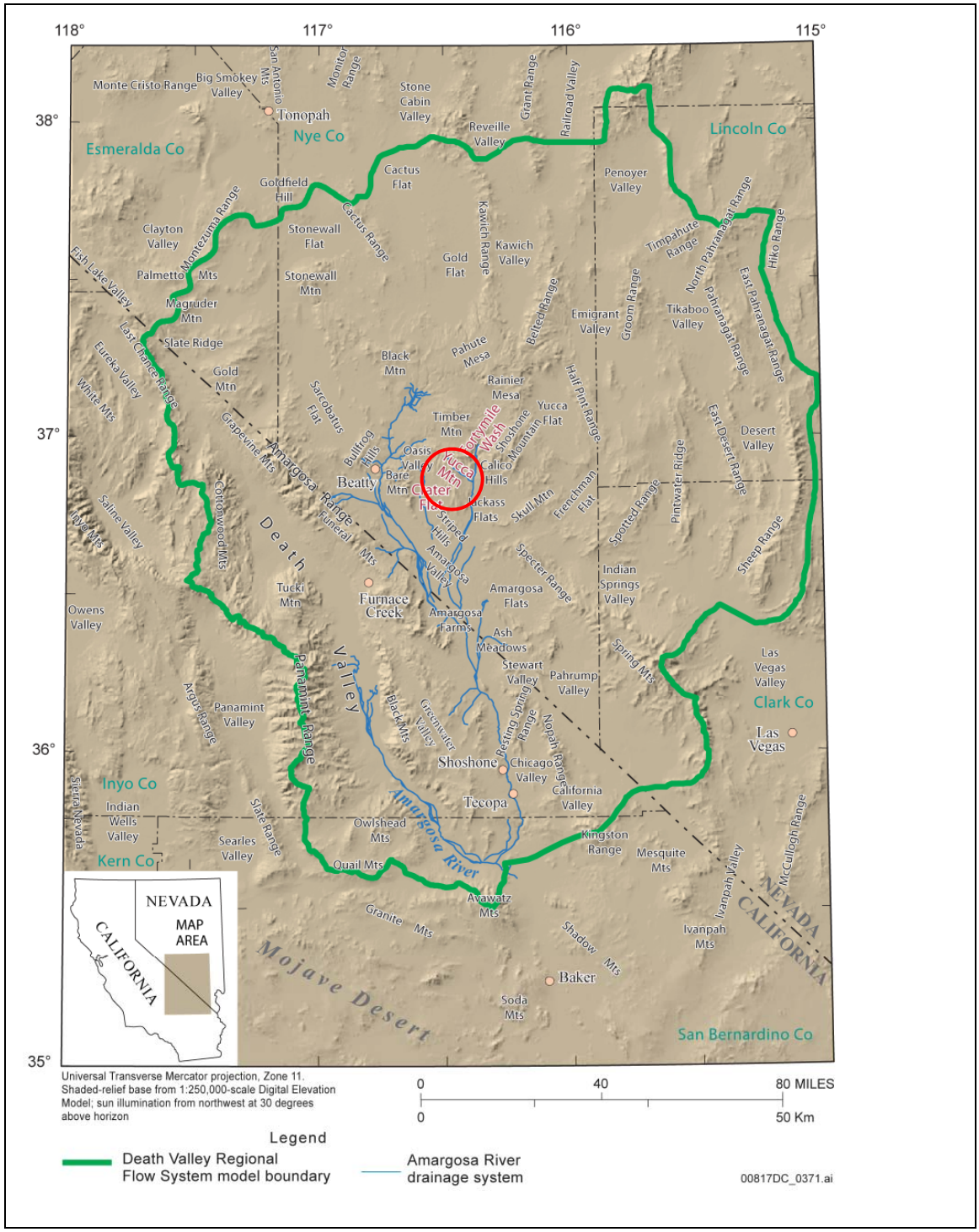

Figure 4. Features of the Death Valley Region (USDOE, 2008, V1 Figure ES-11).

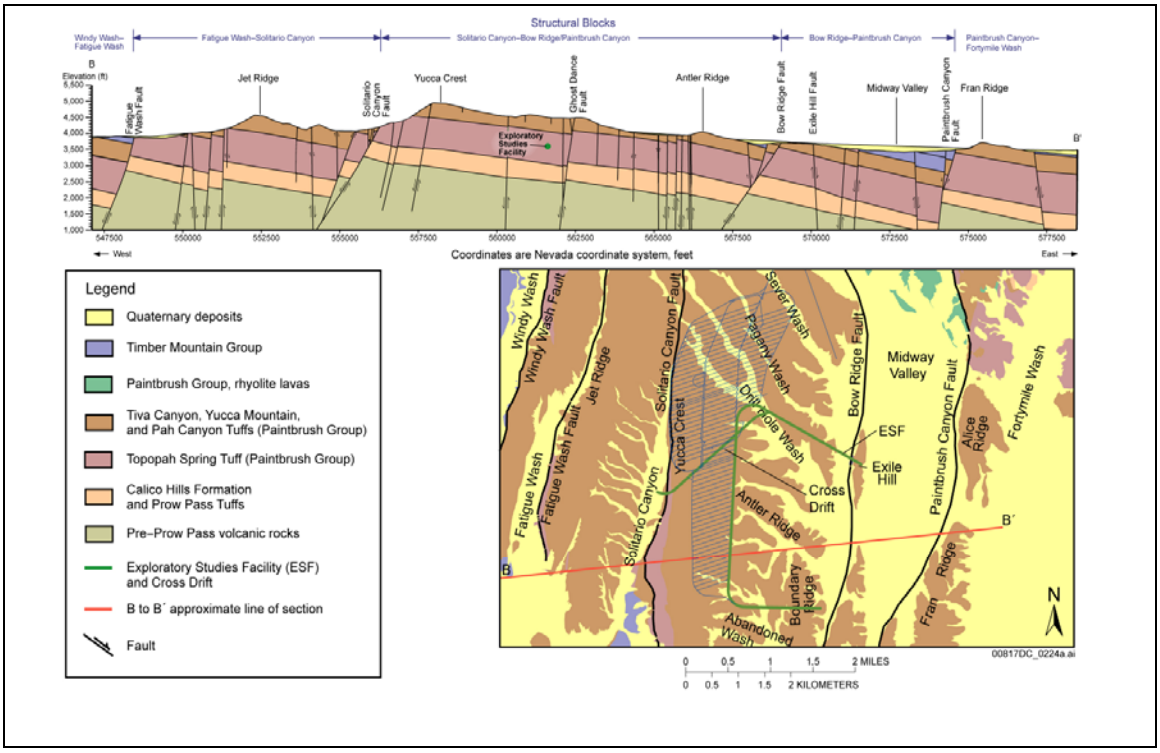

Figure 5. East-west cross-section of Yucca Mountain (USDOE, 2008, V1pg98).

The tectonic setting of Yucca Mountain in the Great Basin is extensional and consists of fault-bonded basins and mountains that over the past 15 million years have been affected and modified by volcanic activity. Strike-slip and normal faults are typically found in the Great Basin. These faults have extensional deformations caused by the plate tectonic interactions of the North American continent. The structural geology surrounding Yucca Mountain is predominately north-trending normal faults with movement down and to the west, some of which show evidence of quaternary movement (i.e., within the last 1.8 million years) (USDOE, 2008).

Two types of volcanism have occurred in Yucca Mountain area, silicic and basaltic. Early phase Miocene silicic volcanism occurred with the eruption of four

voluminous ash-flow tuffs (Sawyer et al., 1994). Yucca Mountain area is composed of uplifted, remnants of these ash-flow tuff deposits (USDOE, 2008). During the Quaternary Period volcanism continued with small-volume basaltic eruptions.

According to the TSPA the 15 million year history of the volcanism in the region peaked between 11 and 13 million years ago and is considered one of the least active volcanic areas in the western United States. 99.9 percent of the volcanic eruptions occurred before 7.5 million years ago. Of the remaining 0.1 percent there has been a 2.5 million year time interval between eruptions. The Miocene eruptions occurred approximately between 9 and 7.3 million years ago and post-Miocene eruptions occurred approximately between 4.8 and 0.08 million years ago. This is the longest break in basaltic eruptive activity in the Yucca Mountain region during the past 9 million years (USDOE, 2008).

3.1.2 Hydrology

Yucca Mountain is situated in the Amargosa River drainage basin, where stream-flow from Yucca Mountain is captured by drainage into the Amargosa River (see Figure 5). The Amargosa River and its tributaries are ephemeral streams; they are dry most of the year with water-flow only occurring in response to precipitation (USDOE, 2008).

Much of the precipitation that falls on Yucca Mountain will either run off, evaporate, or be taken up by the native plants. The small amount that is left over will infiltrate through the soil and into the rock. The water that infiltrates will naturally move downward through the UZ. This water will provide the flow and transport mechanisms

that will move radionuclides from the proposed repository to the SZ. In the SZ the groundwater below Yucca Mountain flows south towards discharge areas in the Amargosa Desert (USDOE, 2008).

Figure 6 shows water movement through Yucca Mountain. Precipitation is the main water input. Once precipitation reaches the ground surface, it percolates through

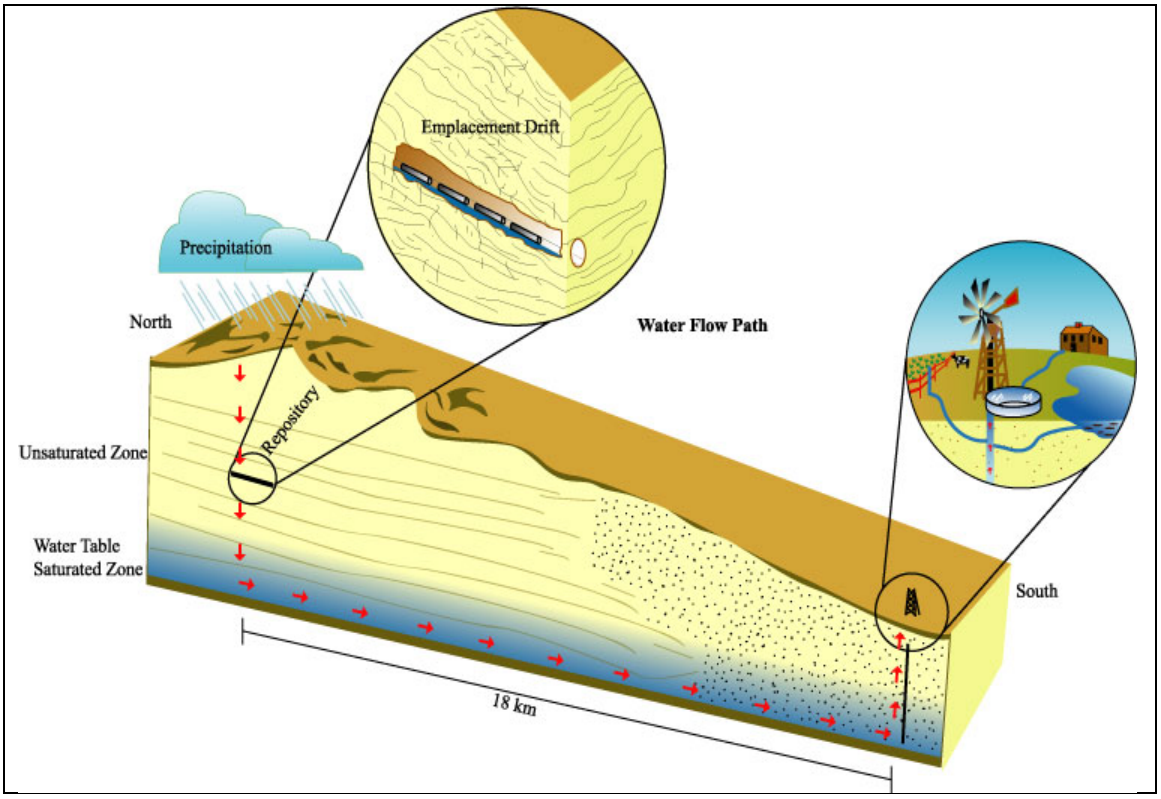

Figure 6. Water flow through Yucca Mountain.

Adapted from V1p109 of the United States Department of Energy (USDOE), 2008, Total System Performance Assessment (TSPA) Model Analysis for the License Application, Volume I, II, III and Addendum 01, Yucca Mountain Project, Sandia National Laboratories, DE-AC04-94AL8500.

the UZ until it encounters the repository located about 300m below the ground surface. The repository will be the starting point for potential radionuclide transport. From the repository, water will continue downward through the rock matrix until it encounters the SZ, 300 meters below the repository. The SZ will then carry the potentially contaminated water in the direction of the groundwater movement. According to the TSPA, human risk calculations are done at the compliance boundary (18km) where groundwater could be pumped out of the ground and consumed (USDOE, 2008).

3.1.3 Geochemistry

The geochemistry of the fluids and rocks in the natural system surrounding the repository will greatly influence the radionuclide transport. The majority of the transport in the welded tuff will be through fracture flow. Advective transport can be defined as the movement of dissolved or colloidal materials within a fluid. Advection of water through fracture networks is expected to dominate radionuclide transport. Therefore, it is important to examine the mineralogy of the fillings and coatings in the fractures to help determine the retardation factors. The rock matrix in the UZ is composed of mainly feldspars and silica minerals (USDOE, 2008). The fracture coatings in the UZ include clays, zeolites, manganese oxides, calcite, amorphous silica (opals), and iron oxides, all of which can increase the sorption factor, therefore increasing the retardation of each radionuclide (BSC, 2004).

3.1.4 Unsaturated Zone (UZ)

Two regions, areas that are and are not underlain by zeolitic rock, represent the UZ. The UZ includes three layers (the Prow Pass, Calico Hills, and Topopah Spring

units) (Figure 5) and UZ is defined by two boundaries: the upper boundary is the ground surface and the lower boundary is the SZ (SNL, 2008; USDOE, 2008). The UZ rock is composed of fractured media globally connected across all cells sharing common interfaces (SNL, 2007a). Within the UZ the transport of radionuclides is expected to be primarily by advection of dissolved species including colloid-facilitated transport as well as matrix diffusion. The TSPA considers colloids to be fine particles ranging in size from 1 nm to 1 μm. These colloids are small enough to be suspended in liquid; radionuclides can be sorbed onto the colloids and will therefore become potentially mobile. Radionuclides sorbed onto mobile colloids could enhance transport rates by allowing transport along fractures in the rock matrix (USDOE, 2008).

The groundwater flow rate, the porosity of the media, interactions between aquifer materials, and the colloidal or dissolved radionuclide's will determine the extent of the advective transport. The porosity of each rock type allows some radionuclides to travel faster (higher porosity vitric rock) or slower (lower porosity zeolitic rock) than the average advective transport time. The porosity of each rock type also affects dispersive processes. Porosity values in the TSPA ranged from 0.01- 1 depending on the scenario model: with average porosity values of 0.3 for alluvium rock and 0.22 for the volcanic rock units (USDOE, 2008).

3.1.5 Saturated Zone (SZ)

The SZ is divided into two segments: (1) fractured-volcanic rock extending from beneath the EBS to 15km down gradient, and (2) alluvium extending from 15km down gradient to the location of the RMEI 18km down gradient (USDOE, 2008). When

radionuclides exit the UZ they enter the SZ where water movement is lateral. The transport continues down the flow gradient to and beyond the compliance boundary at 18 km.

The groundwater flow processes (matrix diffusion, sorption, retardation factors, and radioactive decay) determine the rate of the water movement in the SZ. Radionuclides that reach the SZ will be subject to transport, south, in the groundwater flow direction. The SZ rock matrix, volcanic tuff and alluvium, will determine the fracture flow path the groundwater is likely to travel (USDOE, 2008).

The time for radionuclides to reach any specified point in the SZ down gradient from the repository, such as the compliance boundary (Chapter II), will depend primarily on groundwater velocity. However, matrix diffusion and retardation factors of radionuclides by sorption on the mineral surfaces within bedrock or alluvial aquifers will also affect their transport rate.

3.1.6 Matrix Diffusion

Diffusion from water in rock matrix fractures will retard dissolved radionuclide transport. The diffusion effectiveness of the matrix with regards to retarding radionuclide transport will depend on the diffusive properties of the matrix and the degree of spacing between the fractures (USDOE, 2008).

3.1.7 Sorption

Dissolved radionuclide aqueous species traveling through the rock matrix can be retarded by sorption on the surfaces of matrix materials (i.e. surface of minerals in the SZ rock matrix) a process that could be potentially important for the performance of the

repository. The TSPA describes sorption as the combination of chemical interactions between solid phase (i.e. rock matrix, colloids) and dissolved solutes (USDOE, 2008). Uranium-238 will be sorbed, to some degree, onto colloids. Some of the colloids bearing radionuclides diffuse into the rock matrix pores and fractures affecting the transport times (SNL, 2008). Radionuclides sorbed onto colloids can be potentially mobile, unlike the sorption of radionuclides onto the rock in the rock matrix that are immobile.

The TSPA uses a linear equilibrium sorption model characterized by a single parameter, the sorption coefficient K_d to estimate the radionuclide sorption on the rock matrix (USDOE, 2008). The TSPA used sorption coefficients that were a function of factors including, the heterogeneity and mineralogy of the rock matrix, and the groundwater aqueous chemistry. Sorption coefficients can be affected by temperature. Elevated temperatures generally cause higher K_d values for cationic species, resulting in longer transport times for each radioactive nuclide (SNL, 2007a).

3.1.8 Retardation Factor

Several retardation factors (R_f) are used to determine the effective transport rate for sorbing species in the TSPA and are derived from the matrix sorption coefficient (K_d) values in conjunction with bulk density, matrix porosity, and matrix saturation values. The stronger the sorption, the longer the transport time will be for each radionuclide (USDOE, 2008).

According to the TSPA, radium-226 exhibits high sorption in the UZ tuff layers, particularly in zeolitic tuff as well as in the SZ volcanic and alluvium units. These sorption properties have a slowing effect on the rate of migration of radium-226, to the

extent that decay can occur decreasing the activity concentration before reaching the compliance boundary (USDOE, 2008).

3.1.9 Radioactive Decay

According to the TSPA the overall effectiveness of the transport mechanisms is dependent on two factors: radioactive decay and sorption/desorption. Radioactive decay leads to changes in radionuclide concentrations. In simple parent-progeny decay, the activity of the parent radionuclide decreases with time. The transport models in the TSPA are complicated by chain decay because of the ingrowth of radioactive daughter isotopes created from the decay of the parent radionuclides (USDOE, 2008).

3.1.10 DOE Transport Model

The transport of radionuclides through the geosphere involves many factors including: source concentrations, aqueous speciation, groundwater flow, colloid capture, matrix-fracture interactions, geochemistry of soil and strata, sorption, and solubility (all discussed earlier in this chapter). The radioactive isotope will be transported by water flow and radionuclide concentrations can also be calculated along the transport path according to a transport equation, which takes into account these factors. The TSPA simulates the release of radionuclides from the repository. This release depends on the following: degradation rates of each component of the EBS, dissolution rates of the waste forms, solubilities of the individual radionuclides, whether or not the released radionuclides are dissolved or attached to colloids, whether or not the radionuclides are sorbed into the WPs, and volume and rate of water flowing through the EBS (USDOE, 2008).

The TSPA model includes variable flow paths, which may affect the direction, distance, and time of radionuclide transport. Variability of the characteristics of the natural environment, along with the factors controlling transport, leads to variability in radionuclide transport. The TSPA model serves as part of the overall DOE strategy to provide the safe disposal of HLW, the model encompasses the following five steps (USDOE, 2008):

1. Define the goals and boundary conditions in accordance with regulations.

2. Develop the design for the repository in accordance with regulatory standards.

3. Identify and evaluate features, events, and processes (FEPs) potentially relevant to the long-term performance of the repository and use relevant FEPs to establish scenario classes.

4. Develop and use the TSPA model to estimate repository performance.

5. Analyze and interpret the results of the TSPA model simulation with respect to the performance measures established by the regulations.

The TSPA model uses the 5 steps, listed above, to calculate annual doses from both an undisturbed and a disturbed HLW disposal system. The disturbances can result from human intrusion, early drip-shield (DS) (an interlocking titanium-alloy barrier designed to reduce the effects of rock fall and seepage dripping on the WPs) and WP failures, igneous intrusions, and seismic activity (USDOE, 2008).

The first step in the development of the TSPA model involved the identification and screening of the relevant FEPs that could affect the performance of the repository. Identified FEPs were used to develop five scenario classes: Nominal Scenario

Class, Early Failure Scenario Class, Igneous Scenario Class, Seismic Scenario Class, and Human Intrusion Scenario Class. These scenario classes span a range of possible FEPs for both expected conditions and disruptive events (USDOE, 2008).

The TSPA defines three repository subsystems as barriers that will help protect the environment from a potential radionuclide release: The Upper Natural Barrier (UNB), the EBS and the Lower Natural Barrier (LNB) (USDOE, 2008). In combination these barriers are expected to provide long-term waste isolation and will act as a barrier from the release and transport of radionuclides. Each barrier whether natural or engineered, will limit water movement through the system.

The UNB features include topography and surface soils of the mountain, unsaturated tuff units above the repository, and rock from which the repository is built (USDOE, 2008). The UNB is the portion of the geological strata that extends from the ground surface to the bottom of the repository.

According to the TSPA the EBS will consist of emplacement drifts (shown in Figure 6) where the waste will be stored. Each emplacement drift will have an approximate capacity of 100 WPs. Emplacement drifts will be excavated tunnels approximately 5.5m in diameter, having varying lengths from 300m to 800m, and will have a uniform spacing of 81m. The drifts will accommodate 70,000 metric tons of HLW (USDOE, 2008).

The EBS is the manmade components of the repository system. The EBS could eventually end up affecting the annual mean dose for a RMEI if the WP and the DS fail (USDOE, 2008). The TSPA uses the following mechanisms in the analysis that could

potentially lead to early DS failure: improper heat treatment of the metal, base metal selection flaws, improper weld filler material, and overall emplacement errors. The TSPA identifies the following mechanisms as potentially leading to early WP failure in the analysis: weld flaws, improper heat treatment and stress relief of the outer corrosion barrier and lid, damage by WP mishandling, and improper weld filler material. This thesis will assume a breach in the EBS has occurred, the DS and WP have both failed and radionuclides are being released at a constant rate into the UZ.

The LNB extends from the bottom of the repository to the SZ. Its features include the volcanic rock beneath the repository in the UZ and the volcanic and alluvial material in the SZ (USDOE, 2008).

According to the TSPA the attributes of the natural and engineered system are: limited amounts of water entering the EBS, lifetimes of WPs and DSs, limited radionuclide mobilization and release from the EBS, and retarded radionuclide transport in natural systems after the release from the EBS (USDOE, 2008). These attributes will result in slower overall transport times. The TSPA breaks transport into three major components: EBS flow and transport, UZ flow and transport, and SZ flow and transport. Each component is described below. This thesis will encompass UZ and SZ flow transport into the calculations but will assume a constant release rate from the EBS.

The EBS flow and transport model component calculates radionuclide release rates from the EBS into the UZ. The calculations are determined by seepage into the emplacement drifts, condensation on the drift walls, WP and DS degradation, the presence of water internally in the WP, waste-form degradation, and the overall

environment of the EBS. This component simulates the rate of water flow through the EBS, diffusive and advective transport, sorption, and colloid-facilitated transport (USDOE, 2008).

As described in the TSPA, the UZ flow model component defines the distribution of water flow from the land surface through the UZ as well as seepage into the EBS. The water at the repository horizon, 300m below the land surface, originates from precipitation, as rainfall and snow. Climate, infiltration, drift-wall condensation, and drift seepage all contribute to the UZ flow. The TSPA also includes an UZ transport model component. This component describes the movement and migration of radionuclides from the EBS through the UZ until the point at which the radionuclides reach the SZ. The UZ transport model component simulates advective, dispersive and diffusive transport. The TSPA notes sorption, colloid retardation, and radioactive decay and ingrowth all can affect UZ transport (USDOE, 2008).

The TSPA SZ flow and transport model component simulates the movement and migration of the radionuclides from the water table to the compliance boundary 18km down gradient from the repository. This component simulates advection, dispersion, and diffusion in factures. The radionuclides in the SZ will be transported as solutes or sorbed to colloids (USDOE, 2008).

The TSPA simulations include a biosphere component in the total transport model, which simulates the transport of the radionuclide into the biosphere resulting in the possible exposure of the RMEI. There are two dominant mechanisms of radionuclide release into the biosphere included in the TSPA: (1) SZ release via groundwater, and (2)

release into the air by ash dispersal resulting from a volcanic eruption (USDOE, 2008). The second mechanism will not be discussed in the thesis. All exposure to the RMEI will be assumed to be through groundwater in this thesis.

The TSPA results show the greatest expected mean annual dose for the first 10,000 years after repository closure is less than 0.23 millirem and the greatest expected mean annual dose for the postclosure period from 10,000 to 1,000,000 years after repository closure is less than 1 millirem. The total expected annual dose values given in the TSPA represents the sum of the expected dose calculations for each of five scenario class modeling cases. The TSPA states that there are uncertainties in the total expected dose resulting from epistemic uncertainty surrounding the repository system (USDOE, 2008).

The RMEI is defined as a person who meets the following criteria: an adult with the similar metabolic and physiological considerations to present day knowledge of adults, lives and uses a well, drilled into the groundwater, above the highest concentration of radionuclides in the area of contamination, drinks two liters of water per day from contaminated wells, and has a diet and lifestyle typical of the current residents in the Amargosa Valley (USDOE, 2008).

The TSPA estimated transport times for radium-226 from the repository to the compliance boundary to be much greater than 10,000 years. (SNL, 2008; USDOE, 2008). The median transport times in the SZ ranged from 18,000 years to more than 1,000,000 years, with a 50^{th} percentile median estimated to be about 731,000 years. With these projected transport times, radium-226 would experience between eleven and up to more

than 600 half-lives of decay before reaching the compliance boundary, thereby reaching low activity concentrations (USDOE, 2008).

Radium-226 becomes important, to the RMEI dose, because of the characteristics of its two precursors: thorium-230 and uranium-234. The radionuclide uranium-234 is transported in water only as a solute while thorium-230 can be transported as a solute or as a reversible colloidal phase. Uranium-234 is weakly sorbed in the geological media therefore it is transported much faster than thorium-230 and radium-226. TSPA estimated transport times for both thorium-230 and uranium-234 and found that the 50^{th} percentile median time was greater than 1,000,000 years for thorium-230 and 8,900 for uranium-234 (USDOE, 2008). These results suggest doses from radium-226 will be direct results of the mobilization and groundwater transport of uranium-234 resulting in chain decay to radium-226. The ingrowth of radium-226 will occur along the groundwater flow path.

The TSPA uses natural analogs, both natural and anthropogenic, to provide a way to compare the repository conditions and materials to observed conditions and materials relevant to the timeframe of the repository. Natural analogs are used for comparison and validation of the model components and to help ensure the TSPA has reasonable results. Analogs will not produce the same results as controlled laboratory or field experiments but can build confidence in conceptual models. They are useful in representing FEPs that could affect the performance of the repository. Natural analog data have contributed to the understanding of numerous aspects of the repository system including: degradation of waste forms, aspects of the EBS, drift stability, seepage, UZ

flow and transport, SZ flow and transport, disruptive events, and the biosphere. The TSPA uses the results of natural analog analyses as input data into submodels of the TSPA. In 1990 the Natural Research Council endorsed the use of natural analogs as natural test cases where geological environments have been subject to naturally occurring radioactive materials for millions of years (USDOE, 2008). In addition to the qualitative results from the natural analogs the TSPA uses two analog sites for performance comparisons: Nopal I uranium ore deposit at Peña Blanca (discussed further in Chapter III, secion 3.2.2) and Cerro Negro volcanic eruption.

The TSPA incorporates into its calculations both assumptions and uncertainties for all the components in order to ensure viable results. The TSPA uses a distribution of values for uncertain parameters, rather than a deterministic or single-value calculation. However, each value used may be important in overall total system performance of the repository.

There are expected uncertainties in the behavior of Yucca Mountain repository system over the next million years. These uncertainties require the TSPA model analyses to be probabilistic in order to encompass the entire range of potential outcomes. According to the TSPA the probabilistic analyses reflect a conservative range of processes, behaviors or parameter values of the inherently variable Yucca Mountain repository system, given that complete knowledge of the system is not attainable. The uncertainties in the TSPA are separated into two categories, the uncertainty is either aleatory: irreducible uncertainty (e.g. future behavior of people), or epistemic: reducible uncertainty (e.g. sorption coefficients, matrix diffusion, solubilities). Aleatory uncertainty

takes into consideration the chance of occurrence of FEPs. There is no amount of exploratory work that will provide a definite answer to whether or not an event will occur at any given time. The TSPA addresses aleatory uncertainties by determining a range of likelihood-of-occurrence for a given timeframe. This range is gathered using various formalized means for combining scientific insights from experts in the field. Epistemic uncertainty deals with the uncertainty in knowledge about a FEP value due to limited data or limited interpretations of the data (USDOE, 2008). In principle, epistemic uncertainty can be reduced by the results from experimental testing and the additional data collection.

There are numerous assumptions made in the TSPA such as: climate change and duration of climate periods both assumes the past periods will be the same in the future, WP's emplacement assumed to be all at the same time, and future RMEI diet assumed to be the same diet as the humans currently residing in the Amargosa Valley (USDOE, 2008).

3.1.11 Albertson Transport Model

The TSPA covers a complex and detailed transport model including many mechanisms as described above. Testing the validity of the TSPA model is difficult because of its complexity. For the purpose of this thesis, a modified and simpler transport model is used to represent the down-gradient transport of radium. The model attempted to incorporate the most important mechanisms including: flow rate in the UZ and SZ, retardation factors, and initial concentration (assumed equal to concentration at the compliance boundary). To evaluate the viability of the simpler transport model, results were compared to the TSPA.

The transport equation (3.1) calculates a change in concentration over time, making use of different diffusion rates, advection rates and beginning source concentrations for radioactive nuclides and can be conceptualized with the following terms: Transport= Diffusion – Advection + Solubility – Sorption – Decay + Ingrowth.

$$\frac{dC}{dt} = D\frac{d^2C}{dx^2} - v\frac{dC}{dx} + \lambda_d\,(C_s - C) - K_d\frac{M_r}{V_w}\frac{dC}{dt} - \lambda_{rd}C_d + \lambda_{ig}C_p \qquad (3.1)$$

$\frac{dC}{dt}$ = change in concentration in the liquid phase with respect to time
$D\frac{d^2C}{dx^2}$ = diffusion term: based on Fick's 2nd law involving the diffusion coefficient (D) (Fick's 2nd law)
$v\frac{dC}{dx}$ = advection term: velocity of water in either zone (v) * concentration gradient
$\lambda_d\,(C_s - C)$ = solubility boundary condition: rate constant of dissolution of radium solid (λ_d) * (solubility limit (C_s) – concentration in the liquid phase(C)).
$K_d\frac{M_r}{V_w}\frac{dC}{dt}$ = sorption term: mass of the rock / volume of water in the rock matrix * sorption coefficient * change in concentration in the liquid phase with respect to time
$\lambda_{rd}C_d$ = decay term: rate constant for radioactive decay (λ_{rd}) * daughter isotope concentration(C_d)
$\lambda_{ig}C_p$= ingrowth term: rate of ingrowth for parent isotope (λ_{ig}) * parent isotope concentration (C_p)

Diffusion can be assumed to be small compared to the water velocity in each zone (advective transport), thus the velocity term will be much greater than the diffusion term. The velocity in each zone is assumed to be constant within that zone. The solubility boundary condition is assumed to be zero because radionuclide aqueous concentrations are expected to exceed the solubility limit, leading to solubility-limited boundary conditions. This assumption was made by the TSPA and was not verified by the activity diagrams obtained from natural analog concentrations in this thesis (Chapter IV, section

4.2). The sorption coefficient is considered constant as water flows through the rock matrix. Both decay and ingrowth were considered zero to make the equation simpler, which was justified by the extremely long half-life for U_{238}. Based on these assumptions, equation (3.1) can be simplified into equation (3.2).

$$D\frac{d^2C}{dx^2} - v\frac{dC}{dx} + R_f\ \frac{dC}{dt} = 0 \qquad (3.2)$$

The diffusion coefficient (D) is small (10^{-5}cm^2/sec). The water velocity or flow rate (v) for the UZ used in the TSPA models ranged from 13 to 64mm/year, with a mean of 32mm/year, which was used in this thesis. The SZ flow rate used in the thesis and in the TSPA was 6m/year (USDOE, 2008). The retardation factor ($R_f = 1 + K_d\frac{M_r}{V_w}$) for U_{238} was 6.78 (unitless). The retardation factor effectively reduces the velocity of transport relative to the velocity of flow (i.e., it produces a retarded velocity of transport). Table 6 shows calculated R_f values for isotopes in the uranium-238 decay series through radon-222. The R_f values were calculated for the thesis and were initially planned on being used in multiple simulations of transport equation but since decay and ingrowth were considered zero, it would have required the use of a more complex transport model. It is interesting to note the calculated R_f value for U_{238} and compare it to the R_f values of its daughters. U_{238} has a relative low R_f value (6.78) compared to Th_{234} (144616.4). These values suggest that Th_{234} will have an extremely retarded flow velocity, which suggests a longer transport time. In contrast, Rn_{222} has an R_f value of 1, suggesting that it will not have a retarded flow.

TABLE 6. CALCULATED THESIS RETARDATION FACTORS

Isotope	K_d (ml/g)	M_r (g/cm^3)	V_w	R_f
U-238	0.4	1.88	0.13	6.784615
Th-234	10000	1.88	0.13	144616.4
Pa-234	5500	1.88	0.13	79539.46
U-234	0.4	1.88	0.13	6.784615
Th-230	10000	1.88	0.13	144616.4
Ra-226	550	1.88	0.13	7954.846
Rn-222	0	1.88	0.13	1

Note: Input values used in the calculations were from the TSPA. $R_f = 1 + K_d \frac{M_r}{V_w}$, where Kd: sorption coefficient. M_r: mass of the rock, V_w: volume of water in the rock matrix (USDOE, 2008).

3.2 Activity Diagrams

Natural analog data was examined for groundwater in environments where uranyl minerals or elevated uranium concentrations exist, given that the majority of radium and radon in Yucca Mountain will come from the decay of uranium (USDOE, 2008). Radium is found naturally in groundwater permitting the comparison of natural data to TSPA modeled data. These data are particularly relevant for radium because of its contribution to the total dose, as shown in Figure 1. Analog data provided values for aqueous concentrations, which were used to model aqueous speciation in Yucca Mountain groundwater using EQ 3/6, an aqueous geochemical modeling software program (Wolery, 1992). The program provided solubility limits for the aqueous concentrations of radium at equilibrium with radium-containing solid compounds (e.g., $RaSO_4$ and $RaCO_3$). $RaSO_4$ and $RaCO_3$ activity diagrams are evaluated in this section.

3.2.1 $RaSO_4$ and $RaCO_3$ Data Modeling

Groundwater data (Appendix A, Table A1) was compiled from the United States Geological Survey (USGS) National Water Quality Assessment (NAWQA) data warehouse and from the literature (Almeida *et al.*, 2004). The USGS wells were located throughout the United States: Idaho, Michigan, Massachusetts, Illinois, and Alaska. While the selected wells encompass a variety of temperate regions and do not match the environmental settings surrounding Yucca Mountain, they were chosen for this thesis strictly on the basis that radium concentrations were reported. Groundwater data was also complied from wells in Regiao dos Lagos, Rio De Janeiro State, Brazil (Almeida *et al.*, 2004). The environmental setting around the wells in Brazil is considerably different than the region surrounding Yucca Mountain, but the TSPA references this study as part of the groundwater data used in the TSPA modeling, justifying its use in this thesis. Data from these wells provided an analog that helped model solubility. Using the compiled data, EQ 3/6 predicted aqueous species concentrations at equilibrium.

Reactions 3.3 and 3.4 below show the dissolution of radium sulfate and radium carbonate minerals, respectively. These equilibrium reactions were used in the calculations performed by EQ 3/6, which assumes that all systems move towards equilibrium. Under standard state conditions, for an aqueous species in an ideal system the activity (a_i) is equal to the molar concentration (m_i). This condition is hypothetical where the activity and molar concentration equal one, which only occurs in pure substances (e.g., water). When dealing with systems under non-standard state conditions, the activities must be calculated by multiplying the molar concentration by an activity

coefficient (γ_I). $a_i = m_i \gamma_i$ where: a_i is the activity, m_i is the molar concentration, and γ_i is the activity coefficient

$$RaSO_{4(s)} \leftrightarrow Ra^{2+}{}_{(aq)+} SO_4{}^{2-}{}_{(aq)} \quad (3.3)$$

$K = 10^{-10.45} = a_{Ra^{2+}} .a_{SO_4{}^{2-}}$ at equilibrium at T, or $\log K = \log a_{Ra^{2+}} + \log a_{SO_4{}^{2-}}$

$$Ra\ CO_{3(s)} \leftrightarrow Ra^{2+}{}_{(aq)} + CO_3{}^{2-}{}_{(aq)} \quad (3.4)$$

$K = 10^{-8.3} = a_{Ra^{2+}} .a_{CO_3{}^{2-}}$ at equilibrium at T, or $\log K = \log a_{Ra^{2+}} + \log a_{CO_3{}^{2-}}$

Values for the equilibrium constants K were obtained from the EQ 3/6 database at 25°C and used to obtain equilibrium activities of both salts, radium carbonate and radium sulfate (EQ 3/6 output files, Appendix A, Table A2) (Faure, 1998). The calculated log activity values were used to compile activity diagrams (Chapter IV, Figures 9 and 10).

3.2.2 TSPA Groundwater Analysis-Analog Sites

The TSPA used two natural analog sites for the groundwater analysis, the 1995 volcanic eruption at Cerro Negro, Nicaragua and the seepage and transport of radionuclides at the Nopal I uranium deposit in the Sierra Peña Blanca in Chihuahua, Mexico. The chemical analysis of these groundwater samples found that radionuclides were generally sorbed or re-precipitated on the rocks surrounding the deposit. The analysis also indicated that there is little UZ transport. The TSPA also suggests that flow can be retarded with only a small percentage of the fracture volume sealed by the re-precipitated minerals. These results from the Nopal I site demonstrate the ability for natural systems to provide a “sink” for radionuclides (USDOE, 2008).

Peña Blanca is a naturally occurring uranium ore deposit that offers the opportunity to examine the groundwater flow and transport of uranium in a climate and geological setting very similar to Yucca Mountain. The ore deposit provides the TSPA with a way to estimate the fate of the waste once it is emplaced in the repository. According to samples taken from boreholes near the ore deposit there is a measurable amount, 300 parts per billion, of uranium in the groundwater 50 m below the ore, however samples analyzed 1km down gradient from the ore deposit found negligible amounts of uranium, suggesting very little down gradient transport. Investigations of the ore deposit indicate that there has been very little transport of radionuclides from the deposit and that the radionuclides that have traveled haven't traveled very far from their sources (USDOE, 2008).

3.2.3 Radon in Groundwater

Aqueous concentrations of radon can be found in groundwater (Almeida *et al.*, 2004) but the TSPA does not model aqueous radon concentrations. This was justified by the fact that radon is a noble gas that will come out of solution and into the surrounding rock matrix, eventually making its way to the surface, or will decay, due to its short half-life (3.8 days), before reaching the surface (USDOE, 2008). Focusing on radium in this thesis was justified by the lack of radon data in the TSPA.

Activity diagrams were not created for radon since the USGS wells examined did not report levels of radon. There are two assumptions one can make on the lack of data for radon they are: (1) concentration levels were very low and not detectable by the

analysis processes or (2) that the USGS simply was not looking for dissolved aqueous concentrations of radon.

The study by Alemedia *et al*., 2004, tested for radon concentrations but found a small percentage of the wells had a detectable concentration. Only two out of the 88 groundwater samples were found with radon concentrations over the minimum detectable limit of (3.0 Bq L^{-1}). The geometric mean concentration of radon noted in the literature was 0.256 Bq L^{-1} with a standard deviation of 7.67 Bq L^{-1}. The noted TSPA studies generally did not test for radon concentrations or did not report findings for radon. Radon was left out of TSPA modeling processes due to its short half-life and its natural gaseous state (USDOE, 2008). The fact that radon is left out of TSPA modeling is concerning and is discussed in Chapter V.

3.3 Risk Assessment

As discussed briefly in Chapter II, humans experience risks from exposure to hazards, every day. Everyone is exposed to hazards from the smallest activity like using a microwave to large hazards like driving a car. Some individuals choose to expose themselves to higher risk hazards, while others avoid hazards. With each hazard there is an associated risk. When dealing with Yucca Mountain the hazard is the possible radiation exposure from the various radionuclides released from the repository.

The perception of risk can be defined as someone's reality, the way he/she views life and that risk (Solvic, 1987). A person might have a perception of the risk of a certain hazard that differs from its true risk. Life experiences and recent events and

popular opinion can distort a person's judgment or perception. Perceptions continuously change with the addition of information and with reactions to media coverage. Risk perception encompasses more than just the public's perception in also takes into account economical and political factors (Solvic, 1987; WHO, 2009).

In order to gain a better perception of risk a person must establish an ethical relationship with it. In the case of radiation, one must gain an education and an emotional attachment with it. There is a stigma that all radiation is harmful to life. The major nuclear disasters like Chernobyl and 3 Mile Island and the atomic bombings in Japan have created this stigma. Society has created a negative connotation with radiation. Establishing a positive connotation with radiation could be the hardest challenge facing the nuclear field today.

In order to create a sense of radiation ethics with society organizations, governments, and the scientific community must provide the tools to educate the mass population about the nuclear industry. The education will require a collaborative effort from scientists, educational institutes, government agencies, independent agencies, and the media. So how does society form an ethical relationship with the nuclear industry? It must start with government leaders establishing an ethical relationship. With so many environmental issues these leaders themselves do not have an ethical relationship with the nuclear industry and therefore do not fully understand it and are not compassionate towards it.

Bottom line issues: Is society against nuclear energy only because of its waste? Or are they afraid of radiation because of the few major radiation disasters? If

society became more educated on both topics would they change their opinion? The principles and concepts associated with radiation risk seem to have gone astray. Society is exposed to ionizing radiation everyday and, as discussed in Chapter II, the exposure comes from outer space, from radionuclides in rocks, air, buildings, and even from within humans themselves, yet society is generally not afraid of buildings, rocks or air. According to the NRC, the average amount of radiation a person living in the United States receives annually is 620 millirems (NCRP, 2009). This statistic most likely doesn't mean anything to society because of lack of education associated with radiation.

A radiation education and communicating risk is just as important as calculating risk. Concerned citizens affected by nuclear waste risks will express their views publicly of the uncertainties that they have. Some of the concerns could be remedied by educating society about the hazard and risk with nuclear waste. Communication and education needs to be done in a clear and effective way in order to change the public's perception of risks associated with nuclear waste.

As to the ethical side of nuclear waste, isn't there an ethical responsibility of the generation who created the waste to dispose of it? Currently there is no accepted solution to the nuclear waste problem. The idea of long-term storage (like the proposed repository) would only be a temporary solution. Long-term storage faces many challenges as discussed throughout the thesis. There are uncertainties involved with the performance of the repository and its ability to contain the radionuclides (USDOE, 2008). There is no guarantee that the repository will still function properly 10,000 years or 1,000,000 years from now.

The purpose of a risk assessment is to give information to decision and policy makers as well as the public about the risks and the consequences of possible actions allowing them to manage risk. Important decisions such as: where to place a waste treatment plant, the remediation of a toxic site, minimizing waste generation, or how to dispose of radioactive waste must be made using risk assessment calculations. Risk assessment involves identification, characterization, and quantification of hazards while risk management involves risk communication, decision-making, and mitigation (WHO, 2009). The focus of this section is to assess the risk associated with radium-226 potentially released from the proposed geological repository at Yucca Mountain.

Risk assessment is a formal process used to help better understand the risks associated with health and environmental hazards. The process includes data collection, data evaluation, exposure assessments as well as risk characterization (USEPA 1989,1991). The National Research Council (NCR, 1983) defines risk assessment as the description of potential adverse health effects of human exposure to environmental hazards.

Listed below are the four common steps involved in completing a risk assessment for a carcinogenic substance. (LaGrega et al., 2001):

1. Hazard identification: in this stage information is gathered and processed on the health effects that may be produced by a substance (chemical or radionuclide) at a site and on the conditions of exposure under which the health effects were produced. Specific discussion in this section and thesis addresses the hazard of radiation exposure from a

breach in the EBS and leaking a WP or many WPs at the proposed repository at Yucca Mountain.

2. Exposure assessment: the second stage is to estimate the potential magnitude of exposure from a specific radionuclide. This step determines the pathways (routes of exposure). This stage will help identify the source of contamination and the spatial and temporal distribution of the radionuclides at the specific site.

3. Dose-response or toxicity assessment: in this stage an evaluation is performed using quantitative information and data for a dose of a specific radionuclide and how it relates/correlates to the occurrence of an adverse health effect based on that dose. This quantitative information can be plotted on a dose-response curve.

4. Risk characterization: this is when the data from exposure and dose-response assessment are reviewed. Qualitative and quantitative conclusions are made about the risk; estimations of the number of deaths and uncertainty in the parameters/calculations are made.

Quantitative risk assessment for radionuclide/radiation risks requires an estimate of the exposure of a population to a radionuclide, e.g., radium-226. The estimation of this exposure is known as the Exposure Assessment (USEPA, 1989). Performing an exposure assessment and evaluating an environmental exposure pathway requires several steps. First there must be a source of contamination (e.g., nuclear waste repository, nuclear power plant, toxic waste dump, etc.). In this thesis the source contamination is the proposed nuclear waste repository at Yucca Mountain. Second, there must be a release of that contamination into the environment. The contamination will

follow a transport pathway (e.g., groundwater flow) until it potentially comes in contact with a specific population (e.g., consumers of groundwater). Next, sensitive populations (e.g., children, elderly, etc.) within a general population must be accounted for and there must be an estimation of both long-term and short-term exposures in terms of dose, by a route of exposure (LaGrega et al., 2001; USEPA, 1989).

Once it has been determined that a contaminant could be released from a source it is important to determine how the contamination will react with the environment and what will happen to the contamination over time. With a release of radionuclides from Yucca Mountain the first concern would be the exposure to the initial radioactive isotopes. However, because some radionuclides are unstable and decay to other unstable isotopes during this process there is a continuous release of radiation (see Chapter II for further discussion). For certain isotopes like uranium-238, with half-lives measured in billions of years, there will be a long-term presence of the radiation. There are other short-lived isotopes in the uranium decay series such as ^{222}Rn that has a half-life of only 3.8 days (see Chapter II for half-life, initial inventory, and ingrowth). The short-lived isotopes pose the greatest risk to society, because once they are consumed by a RMEI they will decay to their daughter product releasing radiation inside the person.

The RMEI must come in contact with a radionuclide in order to preform a risk assessment. The routes of exposure for radionuclides and for the risk assessment part of this thesis are found in Table 7. Groundwater and soil contaminated with radium-226 are the focus of this section.

TABLE 7. ROUTES OF EXPOSURE FOR RADIONUCLIDES (USEPA, 1989)

Medium	Routes of Exposure
Groundwater	Ingestion, dermal contact, inhalation during showering
Surface water	Ingestion, dermal contact, inhalation during showering
Food	Ingestion
Soil	Ingestion, dermal contact, inhalation
Air (indoor and outdoor)	Inhalation of ionized gas (radon), inhalation of particles, dermal contact

The routes of exposure (Table 7) are used to estimate the magnitude of an individual's exposure to a specific radionuclide. Some pathways might not be clearly established, while others are clear. When considering an individual's potential interaction with a specific medium one must examine all routes of exposure (USEPA, 1989).

For the Yucca Mountain nuclear waste repository the potential release of radionuclides into the environment could come from natural processes such as 1) dissolution and groundwater transport, i.e., processes modeled with the transport equation (Chapter III, section 3.1), 2) volcanic eruption and earthquakes, 3) human activity, such as building and construction of the repository (not discussed in this thesis), 4) and from possible accidents before or while the repository is being filled (not discussed in this thesis) (USDOE, 2008).

The release of radionuclides from the proposed repository discussed in this thesis allows the RMEI to be exposed to radium through direct contact with groundwater or through exposure to contaminated soil that has been in contact with groundwater. The

exposure equations examined in this section are for RMEI exposed to radium through residential water (i.e. drinking water or showering) and through exposure to soil (i.e. inhalation of soil particles or direct contact with the soil). The exposure equations allow the calculations of an individual's total lifetime excess cancer risk given a specific radium concentration in water/soil. When dealing with radionuclides, the accepted input quantities are in units of activity (i.e., picocuries, pCi) instead of units of mass (i.e., grams, g).

A general understanding of the Total Risk equation (equation 3.5) used in risk assessment is needed to evaluate the results in Chapter IV, section 4.3. The Total Risk equation is used for calculating the total risk estimate for a RMEI for each medium and route of exposure (USEPA, 2004).

$$Total\ Risk = CDI \times SF \tag{3.5}$$

Where CDI is the chronic daily intake of the radionuclide and SF represents the slope factor. CDI is the intake into the body or exposure of the body to a substance. In this section the substance will be the intake of radium-226. The intake factors in parameters such as how much water an individual consumes on a daily basis, the amount of inhaled air daily, and the amount of time an individual is exposed to the radionuclide. SF's are radionuclide specific and have been calculated for each radionuclide based on their chemical, radiological, and metabolic properties (USEPA, 1991). The dose response model accounts for the absorption into the body through the different routes of exposure in the gastrointestinal tract (ingestion), lung tissue (inhalation), and skin (external

exposure). Age, weight, and sex of individuals at the time of exposure affect the dose response relation. The USEPA's average lifetime exposure is 70 years (USEPA, 1989).

3.3.1 Model Residential Water-Carcinogenic Effects

The residential water exposure equation (equation 3.6) is listed below as well as the parameters, definitions, and default values (Table 8): (USEPA, 1991)

$$TR_{residential\ water} = (RW \times IR_w \times EF \times ED)SF_o + (RW \times K \times IR_a \times EF \times ED)\,SF_i \quad (3.6)$$

TABLE 8. PARAMETERS, DEFINITIONS, AND VALUES USED IN THE TOTAL RISK EQUATION FOR EXPOSURE TO CONTAMINATED WATER (EQUATION 3.6)

Parameter	Definition (units)	Value
RW	radionuclide concentration in water (pCi/L)	0.18pCi/L
TR	target excess individual lifetime cancer risk (unitless)	1.00E-06
SF_i	inhalation slope factor (risk/pCi)	1.16E-08
SF_o	oral (water ingestion) slope factor (risk/pCi)	3.85E-10
EF	exposure frequency (days/year)	350 days/yr
ED	exposure duration (yr)	30 yr
IR_{air}	daily indoor inhalation rate of air (m^3/day)	15 m^3/day
IR_{water}	daily water ingestion rate (L/day)	2 L/day
K	volatilization factor	.0005 x 1000 L/m^3

Exposure in residential water will come primarily from direct ingestion of drinking water; however, there can be exposure from inhalation of volatile radionuclides released from water (e.g., during showering). In this section the total risk (TR) for the water pathway will includes two exposure routes: ingestion rate (IR_{water}) and inhalation

rate (IR_{air}). The ingestion rate of water is assumed to be the average amount of water consumed by the RMEI in one day. The inhalation rate is expressed as the volume of air that a RMEI would inhale in one day. With regards to the inhalation rate there is a volatilization factor K, which is different for each radionuclide (Andelman, 1990). The toxicity information (i.e., inhalation, oral, and external exposure SFs) for radium-226 was obtained from the Radionuclide Table, Radionuclide Carcinogenicity, formerly Health Effects Assessment Summary Tables or HEAST tables (USEPA, 1995). Information found in Table 9 is from the HEAST tables for radium-226. The parameter EF (exposure frequency) represents how often the person is exposed to the radionuclide, in this case expressed in days of exposure per year. The parameter ED (exposure duration) represents to how long the person is exposed to the radionuclide, in this case expressed in years.

TABLE 9. TOXICITY INFORMATION FOR RADIUM

Element (Atomic Number)	Isotope	Radioactive Half-life (yr)	Soil Ingestion Slope Factor (risk/pCi)	Water Ingestion Slope Factor (risk/pCi)	Inhalation Slope Factor (risk/pCi)	External Exposure Slope Factor (risk/yr per pCi/m^2)
Radium (88)	Ra-226	1.60E+03	7.29E-10	3.85E-10	1.16E-08	2.29E-04

Source USEPA HEAST tables (United States Environmental Protection Agency (USEPA), 1995, "Radionuclide Table: Radionuclide Carcinogenicity- Slope Factors" http://www.epa.gov/radiation/heast/index.html)

3.3.2 Model Residential Soil-Carcinogenic Effects

The residential soil exposure equation (equation 3.7) is listed below as well as the parameters, definitions, and default values (Table 10): (USEPA, 1991)

$$TR_{residential\ soil}$$

$$= \left(RS \times EF \times IF_{\frac{soil}{adj}} \times 10^{-3} \frac{g}{mg} \right) SF_o$$

$$+ \left(RS \times ED \times D \times SD \times (1 - S_e) \times T_e \right.$$

$$\left. \times 10^{-3} \frac{g}{mg} \right) SF_e \qquad (3.7)$$

TABLE 10. PARAMETERS, DEFINITIONS, AND VALUES USED IN THE TOTAL RISK EQUATION FOR EXPOSURE TO CONTAMINATED SOIL (EQUATION 3.7)

Parameter	Definition (units)	Value
RS	radionuclide concentration in soil (pCi/g)	5.0pCi/g
TR	target excess individual lifetime cancer risk (unitless)	1.00E-06
SF_o	oral (soil ingestion) slope factor (risk/pCi)	7.29E-10
SF_e	external exposure slope factor (risk/yr per pCi/m^2)	2.29E-04
EF	exposure frequency (days/year)	350 days/yr
ED	exposure duration (yr)	30 yr
$IF_{soil/adj}$	age-adjusted soil ingestion factor (mg-yr/day)	3600 mg-yr/day
D	depth of radionuclides in soil (m)	0.1 m
SD	soil density (kg/m^3)	1.43E3 kg/m^3
S_e	gamma shielding factor (unitless)	0.2
T_e	gamma exposure time factor (unitless)	1

The two routes of exposure for residential soil examined in this thesis (Chapter IV, section 4.3) are: (1) direct ingestion (consumption of particulate matter) and (2) external exposure (dermal contact). When dealing with soil ingestion rates age must be factored into the calculations. There are different ingestion rates for children and

adults; children consume more soil than adults. Calculations for risk from soil are done using a uniform distribution of the radionuclide dispersed over an infinite plane and within a finite soil depth (D) and a specific soil density (SD) (USEPA, 1991). Two additional factors the gamma-shielding factor (S_e) and the gamma exposure factor (T_e) must be considered when calculating external radiation exposure. The S_e is always expressed as a fractional value between 0 and 1 and accounts reduction in exposure to radiation from shielding (e.g., dwellings, structures, engineered barriers or topography of the surrounding area). The shields act as a device for trapping the dust, resulting in less possible dust for an RMEI to come in contact with. According to the USEPA the default value is 0.2 (20% reduction in external exposure due to shielding), which is considered to be a conservative number (USEPA, 1981). The T_e is always expressed as a ratio of the number of hours an individual is exposed to the hours in either a full day or that of a work day depending on the type of land (i.e., residential or industrial). The given default value, for residential, is 1 (24 hours of exposure/ 24 hours in a day), which is a conservative assumption. This value assumes that the individual doesn't leave their residence during that 24-hour period (USEPA 1991). The toxicity information (i.e., inhalation, oral, and external exposure SFs) for radium-226 was obtained from the Radionuclide Table, Radionuclide Carcinogenicity, formerly Health Effects Assessment Summary Tables or HEAST tables. (USEPA, 1995). Information found in Table 9 is from the HEAST tables for radium-226.

Currently at Yucca Mountain there is no residential housing and there are no industrial buildings around the planned facility. In the future there could be industrial

buildings to handle the waste. This thesis used the assumption that in the future there could potentially be residential or industrial use of the land surrounding the proposed repository at Yucca Mountain. The modeling, done by the USEPA and the USDOE for the repository on the future performance of the repository are based on the RMEI living 18 km away, where there is planned development and the water table is relatively close to the ground surface (USDOE, 2008).

CHAPTER IV

RESULTS

4.1 Albertson Transport Model Results

Using the Mathematica computational software program, transport times for uranium-238 were calculated (Wolfram Research, Inc., 2008). Modeling was ran using uranium based on the assumption that as soon as uranium-238 reaches the compliance boundary there will be a presence of all daughter isotopes, including radum-226. Modeling using the isotope uranium-238 alone is justified based off simulations and data found in the TSPA. If one isotope in the uranium-238 decay series is found in a sample of groundwater, it is assumed that all other isotopes in the decay series will be present (USDOE, 2008).

Two modeling runs were performed to estimate transport time in the UZ and the SZ. In the UZ a transport time of 6,400 years was calculated to reach a constant concentration at a depth of 300 meters (Figure 7). The calculated transport time in the SZ was 20,600 years to reach a constant concentration at the compliance boundary 18km down gradient (Figure 8).

A calculated total transport time of 27,000 years was acquired by summing UZ and SZ times, and was compared to the TSPA median transport time for Uranium of 330,000 years (USDOE, 2008). The TSPA median transport time is orders of magnitude

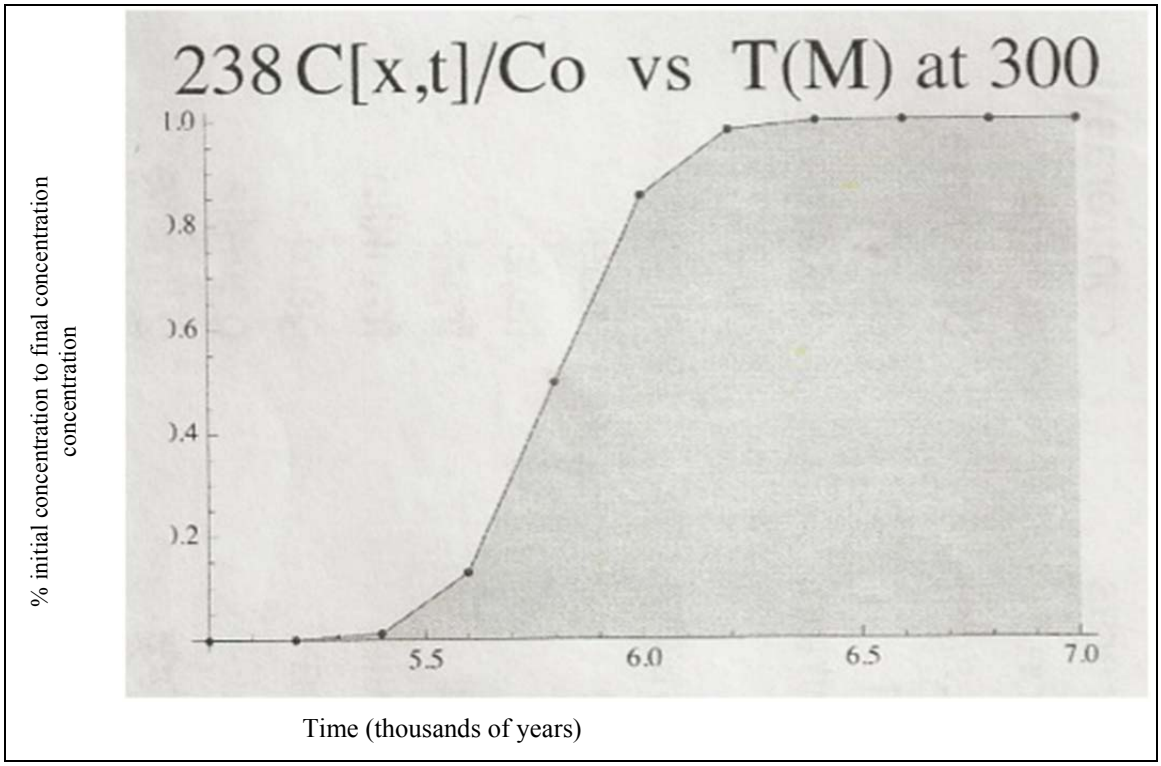

Figure 7. Unsaturated zone transport.

larger than results in this thesis, suggesting that the simplified transport equation used in the model might be overly simplified.

Simplifications that could account for a much faster transport time include: assuming constant flow rates in both UZ and SZ, assuming a constant release rate from the EBS and that the release could maintain the same concentration for the duration of the transport, assuming a constant for any one of the coefficients: mass rock, volume of water, dispersion coefficient and sorbed concentration, and using the mean UZ flow rate in the UZ transport equation. Although this modeling effort produced transport times that were faster than the TSPA, it still constitutes a useful tool to characterize the transport of uranium in Yucca Mountain groundwater. Since the model yielded distinct transport time

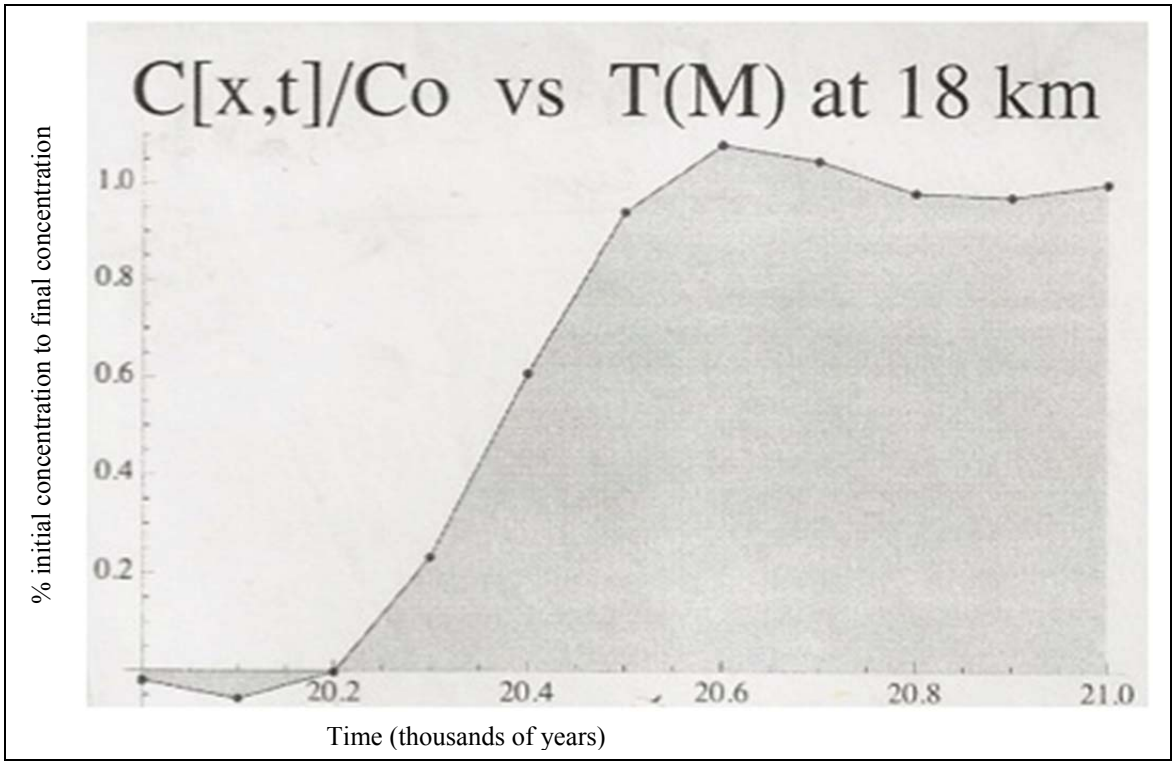

Figure 8. Saturated zone transport.

estimates for the UZ and SZ while relying on the same set of assumptions and simplifications, it appears particularly suitable for comparing transport times between these two hydrogeochemically relevant zones. Chapter V discusses possibilities as to why calculated transport times modeled in this thesis were faster, as well as potential modifications that could be made to account for a transport time closer to the results from the TSPA.

4.2 Activity Diagram Results

Activity diagrams evaluate the status of natural waters relative to the precipitation of radium-containing minerals RaCO3 and RaSO4 (Figures 9 and 10,

respectively). Predicted equilibrium activities of Ra2+, CO32-, and SO42- obtained from the EQ3/6 output results (Appendix A, Table A2) were plotted in log C-log C diagrams and compared to solubility product lines representing dissolution reactions (3.3) and (3.4) (Faure, 1998). The diagrams thus represent solubility limits for RaCO3 (Figure 9) and RaSO4 (Figure 10) in natural waters.

Solubility limits are controlled by many environmental conditions and the TSPA uses these expected conditions, in the repository, when performing solubility calculations. The TSPA calculates solubility limits for a wide range of pH and fugacity (f_{CO2}) values, which control activity coefficient values. Fugacity is an effective pressure (the tendency for a gas to escape or expand) rather than a true pressure. In equilibrium calculations the fugacity of a non-ideal gas is used rather than a true pressure to satisfy the equation for an ideal gas. Fugacity is related to activity by dividing it by a 1 bar therefore affecting activity coefficients. OH^-, CO_3^{2-}, F^-, HPO_4^{2-}, Cl^-, SO_4^{2-}, and NO_3^-, are common ligands found in Yucca Mountain and can form aqueous complexes with radioactive elements (USDOE, 2008). These ligands are included in solubility calculations in the TSPA. Both CO_3^{2-} and SO_4^{2-} were used in the activity diagram calculations in this thesis, since $RaSO_4$ was considered as the controlling radium-containing solid in the TSPA (although the TSPA based radium solubilities off of barium, it is still considered the controlling radium containing solid) and carbonates are generally the most common anion in natural waters. All calculations in this thesis were performed at 25°C.

The modeled activity diagrams for radium carbonate and radium sulfate are shown in Figure 9 and 10 respectively. Data points on both figures represent groundwater chemistry data from Regiao dos Lagos, Rio De Janeiro State, Brazil (Almeida et al., 2004) and USGS NAWQA data warehouse (Appendix A, Table A1).

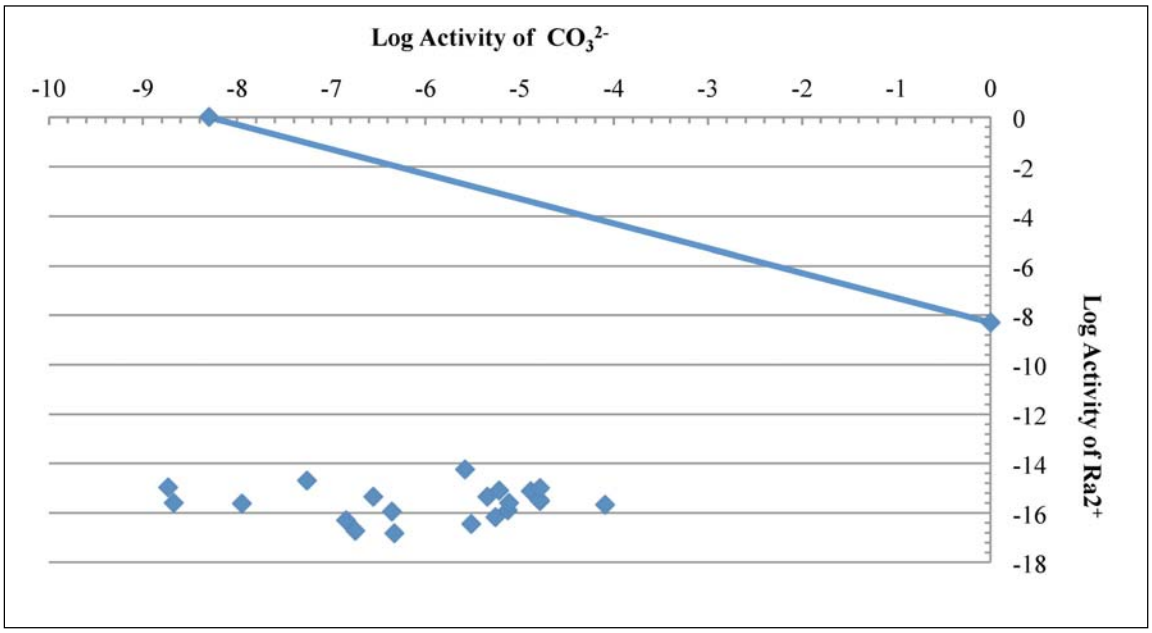

Figure 9. Activity diagram illustrating the stability of radium carbonate ($RaCO_3$) at 25°C and 1 bar. The line represents the solubility product ($K= 10^{-8.3}$) for $RaCO_3$ (reaction 3.4).

The modeled activity diagrams for radium carbonate and radium sulfate are shown in Figure 9 and 10 respectively. Data points on both figures represent groundwater chemistry data from Regiao dos Lagos, Rio De Janeiro State, Brazil (Almeida et al., 2004) and USGS NAWQA data warehouse (Appendix A, Table A1).

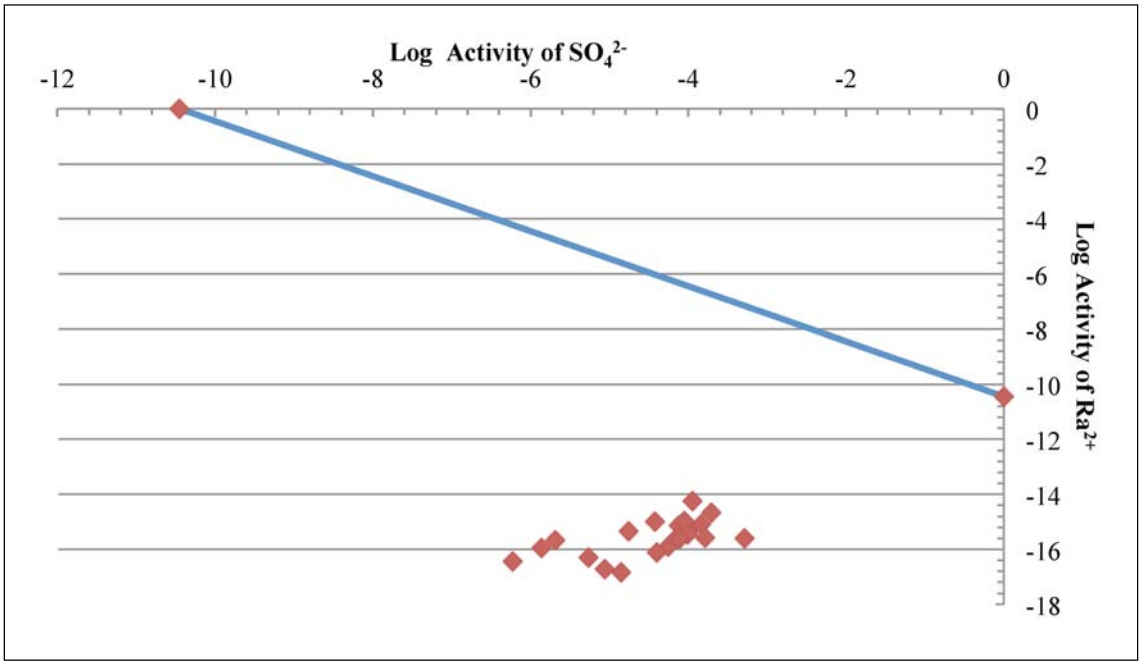

Figure 10. Activity diagram illustrating the stability of radium sulfate ($RaSO_4$) at 25°C and 1 bar. The line represents the solubility product ($K= 10^{-10.45}$) for $RaSO_4$ (reaction 3.3)

The line in each activity diagram shows the equilibrium threshold between saturated and undersaturated conditions, with respect to the salt considered, and was obtained from thermodynamic data. This equilibrium line puts an upper limit on the ionic constituents concentrations of each salt in an aqueous environment. Once the product of ionic concentrations reach this limit, radium will precipitate out as a salt and will therefore come out of solution and will no longer be transported with groundwater down gradient. According to the activity diagrams (Figures 9 and 10), radium sulfate ($RaSO_{4\,(s)}$) and radium carbonate ($RaCO_{3\,(s)}$) are not forming in natural systems, most likely because dissolved ion concentrations will not reach the solubility limits represented by the lines in the figures.

All data points analyzed for both salts fell below the line in the undersaturated zone. This means that currently if radium enters the system as an ion at concentrations similar to natural analog concentrations, it would remain in the ionic form and would not be precipitated out as a salt, therefore radium ions would be carried in water down gradient. These results, that natural systems are undersaturated with respect to $RaCO_3$ and $RaSO_4$, are different than what was expected when comparing to the modeled TSPA results and would not have allowed the solubility boundary condition ($\lambda_d\ (C_s - C)$) to be zero in the transport equation (equation 3.1). A discussion of possible reasons for this discrepancy is provided below.

4.2.1 Activity Diagram Uncertainties and Assumptions

The TSPA modeling suggests that once the EBS fails and radioactive isotopes are released into the environment they won't travel far from the source. However, the results in the thesis suggest that the isotopes will travel further from the repository than suggested in the TSPA.

As discussed in this chapter, the activity diagrams obtained for the natural analog systems suggested that radium would not be precipitated out as a salt ($RaCO_3$ or $RaSO_4$) and would therefore stay in solution and transport down gradient as a cation. This result was different than what was expected. According to the TSPA, radium should be sorbed or precipitated out as a salt and should not be transported down gradient (USDOE, 2008). There could be numerous reasons why this thesis results were different than the TSPA results including: the initial concentration of radium used in TSPA modeling

compared to the natural radium concentrations used in the modeling in this thesis, differences in the temperatures used in modeling, TSPA modeling was done using barium compounds rather than radium compound, and differences in the overall groundwater chemistry of the wells analyzed compared to Yucca Mountain groundwater used in the TSPA modeling.

Differences in initial concentrations of radium considered in modeling likely account for discrepancies in modeled solubility conditions. This thesis used natural groundwater concentrations data, which could have greatly differed from the concentrations used in the TSPA. The TSPA likely assumed greater concentrations of radium in the event of WP failure. Assuming larger initial aqueous concentrations for radium would lead to supersaturated conditions and justify the solubility boundary conditions hypothesized in Chapter III, section 3.1.

Radium solubility for the TSPA is based on calculated barium solubility at 100□C using $BaSO_4$ as the solubility-controlling phase, which is considered a good analog for modeling radium solubility (SNL, 2007b; USDOE, 2008). In this thesis, $RaSO_4$ and $RaCO_3$ were used as natural analogs for modeling and calculations were performed at 25°C. Since barium calculations were performed at 100°C and radium calculation were performed at 25°C, this could be one reason the results from this thesis were different than what was expected. Running thesis models at a higher temperature would have increased the solubility product (K) changing where the line would have fallen on the activity diagrams: which would have led the natural systems more

undersaturated. Ruling out modeling temperature as a reason for the differences in results between the TSPA and this thesis.

The fact that two different solids (radium compounds and barium compounds) are being compared could have attributed to different results than expected. Barium salts and radium salts would have distinct solubility constants; which would affect saturation boundary conditions. The TSPA uses $RaSO_4$ as the controlling solid for radium. Natural analog data shows radium concentrations in some natural waters are orders of magnitude below levels corresponding to $RaCO_3$ and $RaSO_4$ saturation (Figure 9 and 10 respectively). According to SNL 2007b and USDOE 2008 $SrSO_4$ and $BaSO_4$ are more common sulfate solids than $RaSO_4$. Since the TSPA calculates radium solubility based on Ba solubility and uses $BaSO_4$ as the solubility-controlling phase, it gives an overestimation of the aqueous radium concentrations. The TSPA also suggests the water chemistry inside the repository will be similar to the normal range of natural waters therefore normal radium solubility's are acceptable in modeling (USDOE, 2008).

Overall differences in the chemical composition of groundwater in the wells as compared to the Yucca Mountain groundwater used in the modeling of the TSPA, could have also affected the results from this thesis. Thermodynamic speciation models (like EQ 3/6 and the one used in TSPA) take into account the overall composition of water samples. Presence of other ions will affect solubility predictions. Therefore affecting overall modeling results from EQ 3/6.

There are uncertainties as discussed in Chapter III when predicting the concentration of radium at any point along the transport path. One uncertainty is the

length of time required for a radioactive isotope to penetrate the groundwater and flow down the gradient and enter the biosphere. As radionuclides are captured or sorbed onto the rock matrix the concentration will decrease with time as discussed in Chapter III. As ingrowth occurs the concentration will increase over time. Each transport mechanism will contribute in a positive or negative way with respect to the time it takes to transports that isotope down the gradient and the concentrations at the compliance boundary (Chapter III).

Assuming the thesis results should have been the same as the TSPA allowed the solubility boundary condition term, $(\lambda_d\,(C_s - C))$, to be assumed equal to zero (equation 3.1). The solubility boundary condition term λ_d is equal to the rate constant of dissolution of radium solid (λ_d) multiplied by the solubility limit term $(C_s - C)$, where C_s is the equilibrium concentration of radium and C its actual concentration. This assumption allowed the solubility boundary condition $(\lambda_d\,(C_s - C))$ to be zero in the transport equation (equation 3.1), which in turn led to the use of a simplified transport equation in the thesis modeling (equation 3.2).

4.3 Risk Assessment Results

Below are two risk assessment scenarios calculating a total risk using different radium concentrations based on the scenario. Scenario 1 uses natural concentrations of radium in groundwater obtained from the United States Geological Survey (USGS) National Water Quality Assessment (NAWQA) data warehouse (Appendix A, Table A1). The residential water exposure equation (equation 3.6) discussed in Chapter III (section

3.3) is used to calculate a total risk for water exposure. Scenario 1 uses a mean radium groundwater concentration of 0.18 pCi/L; assuming external exposure risks are negligible. The residential soil exposure equation (equation 3.7) discussed in Chapter III (section 3.3) is used to calculate a total risk for soil exposure. Scenario 2 uses the USEPA standard for maximum soil concentration for radium of 5.0 pCi/g in the first 15 centimeters a total risk is calculated; assuming water exposure risks are negligible (USEPA, 2009).

Scenario 1: Residential Water

$$TR_{residential\ water} = (RW \times IR_w \times EF \times ED)SF_o + (RW \times K \times IR_a \times EF \times ED)\,SF_i \quad (4.1)$$

$$= \left(0.18\frac{pCi}{L} \times 2\frac{L}{day} \times 350\frac{days}{year} \times 30\ years\right) \times 3.85^{-10}\frac{risk}{pCi} + \left(0.18\frac{pCi}{L} \times 2\frac{L}{day} \times 350\frac{days}{year} \times 30 years\right) \times 1.16^{-8}\frac{risk}{pCi}$$

$$= 1.4553^{-6} + 1.6443^{-4}$$

$$= 1.6588^{-4}\ x\ 10^{6}$$

$$= 165\ people\ out\ of\ 1\ million$$

The result above shows that if individuals consume two liters of water with a concentration of 0.18 pCi per liter of radium-226 for 350 days of every year for 30 years, 165 deaths out of 1 million would occur from cancer directly related to the radium exposure. The same calculation (equation 4.1) was performed using the USEPA maximum radium-226 groundwater concentration limit of 5.0 pCi/L resulting in 4607 deaths out of 1 million occurring from cancer directly related to the radium exposure (USEPA, 2009).

Scenario 2: Residential Soil

$$TR_{residential\ soil} = \left(RS \times EF \times IF_{\frac{soil}{adj}} \times 10^{-3}\frac{g}{mg}\right)SF_o + \left(RS \times ED \times D \times SD \times (1 - S_e) \times T_e \times 10^{-3}\frac{g}{mg}\right)SF_e \quad (4.2)$$

$$= \left(5.0\frac{pCi}{g} \times 350\frac{days}{year} \times 3600\frac{mg\ year}{day} \times 10^{-3}\frac{g}{mg}\right) \times 7.29^{-10}\frac{risk}{pCi} + \left(5.0\frac{pCi}{g} \times 30\ years \times 0.1\ m \times 1.43^{3}\frac{kg}{m^3} \times (1 - 0.2) \times 1 \times 10^{3}\frac{g}{mg}\right) \times 2.29^{-8}\frac{riskm^2}{year\ pCi}$$

$$= 6300pCi \times 7.29^{-10}\frac{risk}{pCi} + 1.71^{7}\frac{pCi}{year} \times 2.29^{-8}\frac{riskm^2}{year\ pCi}$$

$$= 4.59^{-6} + 0.3915$$

$$= 0.3915\ x\ 10^{-6}$$

$$= 0.3915\ people\ out\ of\ 1\ million$$

This means that if individuals reside in an area with a soil concentration of 5.0 pCi/g of radium-226 in the first 10 centimeters of soil, live there for 30 years and of those 30 years spend 350 day a year at that residence than 0.3915 people out of 1 million will die from cancer related to radium exposure (USEPA, 2009).

4.3.1 Risk Assessment Limitations and Assumptions

When predicting future risk the calculations are only as good as the models and original estimates of beginning concentrations. There are uncertainties at every step. There are assumptions made about frequency and duration of exposure. If the "average person" A is exposed to X their body might react a certain way to such substance yet person B might not react the same way. An average gives the risk assessor the best idea

of what could occur. Scenario 1 assumes that the RMEI's are never exposed to contaminated soil over a 30 year period; this could be unrealistic. If the water is contaminated that the RMEI is consuming on a daily basis then there is a chance that the soil surrounding the site of water consumption could be contaminated as well due to irrigation. A more realistic approach would require a different risk equation or a combination of equations that would factor both contaminated water and soil into it. Scenario 2 assumes that the RMEI's are never exposed to contaminated water over a 30 year period; this too could be unrealistic. If the RMEI is exposed to contaminated soil there is also a chance that the water they are exposed to everyday could be contaminated. A more realistic approach would require a different equation or combination of equations as stated above.

CHAPTER V

CONCLUSIONS AND DISCUSSIONS

The overall objective of this thesis was to grasp a better understanding of the long-term dose risk from radium associated with the proposed high-level nuclear waste repository at Yucca Mountain, Nevada and to test the Yucca Mountain risk modeling by using natural groundwater levels to simulate a potential release of radioactivity. Overall conclusions, questions answered, and a general discussion of results are provided in this chapter.

- *Are the natural analog data and simple transport models realistic or conservative by comparison to the DOE models?*

This thesis encompassed unsaturated zone (UZ) and saturated zone (SZ) flow transport but assumed a constant release rate from the engineered barrier system (EBS). This is most likely not realistic. In reality, if one waste package (WP) failed there would be a finite source of radionuclides that could be released. The release time would vary based on numerous factors such as size and number of the breaches to the EBS, the size of the breach in the WP, as well as environmental conditions surrounding the WP and EBS. The assumption of constant release rate simplified the calculations and made the modeling easier by not having to factor in a change in initial concentration over time.

The time for radionuclides to reach any specified point in the SZ down gradient from the repository, such as the compliance boundary (Chapter III), will depend primarily

on the groundwater velocity, however matrix diffusion and retardation factors of radionuclides by sorption on the mineral surfaces within the bedrock or alluvial aquifers will also affect transport rate. As discussed in Chapter III all of these factors are taken into consideration in the thesis modeling. A simplified transport model was used in this thesis to obtain transport time results. This is justified in the TSPA. The TSPA overall results were compared to a stand-alone simplified TSPA analysis of the Yucca Mountain repository. The simplification involved removing details from the TSPA model to capture spatial and temporal variability and treating the repository system using an average representation of processes and parameter values. Although the simplified TSPA analysis is different than the results obtained from the overall TSPA model in both its structure and computational method, the simplified model has identical technical bases. The TSPA simplified model was still more complicated than the model used in this thesis, since it included seismic and igneous intrusion modeling cases, WP failure modeling, and multiple realizations. In the simplified TSPA model, total mean annual dose and mean annual dose from individual radionuclides were similar in magnitude to those obtained for the corresponding modeling cases using the overall TSPA model, suggesting that a simple model can produce similar results to a complicate multiple step model (USDOE, 2008). These results help support the validity of the simple transport model used in this thesis.

- *What is the transport time for radium-226 and radon-222 to enter the biosphere once the WP and EBS fail?*

The transport times found in this thesis did not break down individual isotopes but rather made the assumption used in the TSPA: that if one isotope in the uranium-238

decay series was found in a groundwater sample then all other isotopes in the series should be present.

As discussed in Chapter III the transport equations solved for a total transport time through both the UZ and SZ. In the UZ, a transport time of 6,400 years was estimated for the final concentration to match the initial concentration 300 meters away from the source. In the SZ, it would take 20,600 years for the final concentration to match the initial concentration 18 km down gradient. A total transport time of 27,000 years was calculated for radioactive waste to travel through the UZ and SZ and reach the drinking water source of a RMEI. This result is a much faster transport time than reported in the TSPA, where a median transport time for uranium was 330,000 years. The TSPA reported a median transport time, while this thesis only modeled transport time with one set of conditions and therefore does not yield a median transport time. If additional models in this thesis would have been run adjusting different factors (i.e., diffusion rate, water velocity, and retardation factors) a median transport time could have been calculated. Comparing the median transport time of the TSPA to this thesis was considered valid since this thesis assumes that the EBS and WP's fail as soon as they are placed inside the repository; these assumptions likely would not happen in reality. In reality the WP and EBS have been designed to last for many millennia. Also, adjustments could have been made in the assumption of length of time before the WP and EBS fail. This thesis considered an immediate WP/EBS failure, with no lag time built into the transport model. Taking the estimated WP/EBS fail time of 300,000 years from the TSPA into account makes the transport time result of 27,000 years comparable to TSPA estimates. In the TSPA, simplified model results showed that under normal conditions a

WP failure would occur on average 300,000 years after emplacement. In the overall TSPA results, WP failure on average occurred as early 100,000 years after emplacement (USDOE, 2008). By adding the transport time calculated in this thesis (27,000 years) to the average WP failure time of 300,000 years, a total transport time from emplacement to the compliance boundary of 327,000 years is obtained for uranium-238, which is very close to the average transport time given in the TSPA (330,000 year).

There are additional factors that could have affected the transport model used in this thesis, as discussed in Chapter IV. Results of the chemical modeling performed in Chapter IV were opposite of what was expected, and required the assumption of supersaturated conditions in order to simplify the thesis transport model (Chapter III). This assumption allowed the solubility boundary condition term, ($\lambda_d\ (C_s - C)$), to be zero, which could have affected the overall transport time results. Not factoring in ingrowth and decay into the transport model could also constitute significant simplifications. Any slight change in water velocity, porosity, temperature, or chemical composition of the water and of the rock matrix could have also affected the transport time estimate.

In regards to radium transport times, additional transport models could have been run in this thesis but would have required ingrowth and decay to be factored into the calculation, resulting in a more complex transport equation similar to equation 3.1. The idea behind the transport equation in this thesis was to run the simplest version of the complex multi-step process used in the TSPA and to compare total transport times to the TSPA results. The TSPA calculated a median transport time for radium of 730,000 years (USDOE, 2008). It would not be appropriate to compare this estimate for radium to this

thesis result for uranium, since they are two different isotopes in the uranium-238 decay series. However a total transport time of 730,000 years is plausible due to the half-lives involved from uranium-238 to radium-226. As discussed in Chapter II, each isotope in the uranium-238 series has a different half-life; each step would require time for decay resulting in a longer transport time for each isotope down the decay series from uranium-238. Potential causes for increased transport times for radium-226 are also discussed in Chapter IV. Radon transport times were not evaluated by the TSPA, therefore radon was not considered in this thesis transport model either.

- *Is the post-closure individual protection standard of 350mRem/year between 10,000 years and 1,000,000 years a realistic number for the YMP?*

Ra-226 is not expected to dominate the mean annual dose until 10,000 to 100,000 years after the repository closure, but will then dominate the mean annual dose for the post-closure period from 100,000 to 1,000,000 years (Figure 1, USDOE, 2008). The results provided by the TSPA include calculations of the total mean annual dose to the RMEI in any year during the next 10,000 years after repository closure and the total median annual dose to the RMEI in any year from 10,000 years after repository closure through 1,000,000 years. The post-closure individual protection standard is set at 15 millirem up to 10,000 years and 350 millirem after 10,000 years up to 1,000,000 years. The TSPA results show that the highest projected total mean and total median annual doses to the RMEI are estimated to be about 0.23 millirem and 0.99 millirem. These results suggest that the highest projected dose values throughout the next 1,000,000 years are more than two orders of magnitude below the individual protection limit of 350 millirem per year (USDOE, 2008). The TSPA results demonstrate that the highest

calculated mean dose to the RMEI in any year during the 1,000,000 years after the proposed repository closes is less than the individual protection standard of 350milirems.

However the standard of 350 millirems itself is of concern. When the standard was set in 2008 the average radiation a person living in the United States received per year was 360 millirems (NRC, 2001b). This made the acceptable amount of possible radiation released from the proposed repository almost the same level that individuals, living in the United States, are exposed to annually. Although the estimated amounts of released radiation from the repository are magnitudes below the standard it is concerning that the allowable amount (350 millirems) would almost match the average background radiation (360 millirems). Since the TSPA results have been released the average amount of background radiation a person living in the United States has been updated to 620 millirems (NCRP, 2009).

This thesis did not calculate annual dosages to the RMEI released from the proposed repository, but it is important to discuss this topic. Since health concerns are a major issue surrounding nuclear waste, it is important to discuss the protection standards set for Yucca Mountain. Figure 1 was the inspiration in starting this study; it shows that the majority of the mean annual dose will come from radium-226. This result drove the overall research for radium-226, the transport equation, and the health effects caused by radium. This result also brought up unanswered questions as to why radon-222 wasn't evaluated in the TSPA since it accounts for such a large amount of the annual dose an individual living in the United States receives yearly.

- *How accurate can the current models predict 1,000,000 years from now?*

The TSPA model analysis shows that the repository will, with a high degree of confidence, function in a manner that protects the environment and future human populations. The work done in this thesis did not try to calculate accuracy of the model but rather discuss the limitations and assumptions used in the TSPA modeling and modeling done in this thesis. The TSPA model was designed to establish confidence that the calculated results were achieved properly using the modeling tools, submodels, and a given set of controlled inputs. There is uncertainty incorporated into the input data and submodel performances, of the TSPA, making the validity of the results more realistic. The TSPA results, when possible, were compared with experimental measurements and/or field observations. However, some measurements were impossible to obtain from modeling and could not be calculated. Therefore, a technical review and natural analogs were used to validate the TSPA (USDOE, 2008).

The analyses performed, in the TSPA model, suggest the performance of the proposed repository system will provide a significant protection to groundwater. The results show concentrations in the groundwater will be below the groundwater protection standard, set by the USEPA. The environmental features (i.e., topography, climate, geology, and soil characteristics) at the site of the proposed repository will add in restricting the amount of infiltration of precipitation into the subsurface. The UZ rock characteristics and infiltration rate as well as the placement of the repository within the mountain will further limit the amount of water available to enter the emplacement drifts. Features of the EBS, together with the solubility of radionuclides and the UZ and SZ geology and hydrology, will limit the release and transport of radionuclides into the SZ.

TSPA results indicate that the characteristics of the SZ (i.e., rock, soil, and the hydrologic factors) will add in retarding radionuclide transport and reduce the amount of radionuclide available for possible human contact in the biosphere (USDOE, 2008). Even with the assumptions, limitations, uncertainties, and all the modeling calculations performed for the TSPA it is still difficult to predict what the Earth will be like 1,000,000 years from now making it hard to accurately predict if the TSPA models are correctly accounting for all factors 1,000,000 years from now.

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APPENDIX A

TABLE A1. GROUNDWATER DATA COMPILED FROM THE UNITED STATE GEOLOGICAL SURVEY (USGS) NATIONAL WATER QUALITY ASSESSMENT (NAEQA) DATA WAREHOUSE AND FROM THE LITERATURE (ALMEIDA ET AL., 2004).

Name	ID # USGS Wells	Na^+ (mg/L)	SiO_2 (mg/L)	Ca^{2+} (mg/L)	K^+ (mg/L)	Mg^{2+} (mg/L)	HCO_3^- (mg/L)	Cl^- (mg/L)	NO_3^- (mg/L)
Brazil	Alamedia et al.	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a
Idaho2	480235116483001	3.385	13.69	53.88	2.880	23.15	251.0	2.85	0.9475
Idaho4	480235116510001	3.846	21.93	31.06	1.850	9.966	140.0	1.30	0.7837
Idaho5	480235117012501	5.055	11.89	50.96	3.320	24.65	278.0	1.21	0.1771
Idaho6	481320116261101	25.00	12.79	35.51	1.270	10.21	120.0	0.79	0.1772
Idaho8	480950116324401	5.187	33.09	5.423	0.180	2.770	38.0	0.60	0.8634
Michigan1	454622087114801	9.757	8.82	52.86	3.391	22.02	257.0	6.70	0.2568
Michigan2	460000088235501	1.630	12.00	33.50	0.636	17.70	175.0	9.23	3.6300
Michigan3	460308088130601	2.080	10.30	22.40	1.110	11.00	115.9	0.41	0.2302
Michigan4	462416088191901	2.083	12.57	28.16	2.154	8.804	119.1	0.90	0.2479
Alaska1	602107151160901	5.724	34.01	12.04	3.160	6.011	71.0	4.28	0.4915
Alaska2	602311151123701	5.724	37.28	7.930	1.820	3.678	55.0	3.32	0.1771
Alaska3	602600151084301	5.224	30.44	13.60	2.220	5.624	81.0	3.50	0.1771
Alaska4	603216151085401	127.300	44.35	24.52	12.55	17.84	178.0	213.25	0.1771
Alaska5	603216151114501	359.500	44.42	4.35	9.93	5.01	468.0	495.45	0.1771
Alaska6	603332151110401	3.570	28.66	10.94	1.100	2.137	42.0	6.07	0.2833
Illinois1	401041088242901	20.670	18.24	68.91	1.567	29.46	413.4	0.64	0.2568
Illinois2	401131088255701	23.910	21.15	76.92	1.538	30.33	1.0	0.67	0.2568
Massachusetts1	414541071142501	28.840	9.83	18.28	5.280	3.053	7.0	45.55	48.0800
Massachusetts2	414913071052701	6.1300	12.01	13.49	1.790	5.292	6.0	9.98	36.7300
Massachusetts3	41520071082401	25.900	14.89	12.62	0.780	3.861	17.7	53.28	0.6332

Table 1. (Continued)

Name	SO_4^2 (mg/L)	Ra^{2+} Activity (pCi)	Ra^{2+} (mol/L)	U^{4+} (mg/L)	UO_2^{2+} (mg/L)	pH	E_h (V)	T (ºC)	$\log(a_{CO3^{2-}})$	$\log(a_{SO4^{2-}})$	$\log(a_{Ra^{2+}})$
Brazil	n/a	n/a	2.80E-15	8.35E-05	9.44E-05	n/a	n/a	n/a	-7.260	-3.710	-14.680
Idaho2	15.853	0.140	6.28E-16	3.11E-03	3.51E-03	7.58	7.55	7.6	-5.342	-3.993	-15.351
Idaho4	7.474	0.233	1.61E-16	1.14E-03	1.29E-03	8.05	9.42	7.9	-5.126	-4.260	-15.905
Idaho5	11.817	0.036	1.05E-15	3.98E-03	4.49E-03	8.02	0.03	7.6	-4.881	-4.120	-15.129
Idaho6	73.709	0.077	3.45E-16	1.40	1.47E+00	8.10	0.02	9.0	-5.112	-3.287	-15.596
Idaho8	0.613	0.013	5.83E-17	1.00	1.13E+00	6.84	9.14	8.9	-6.843	-5.266	-16.296
Michigan1	17.585	1.179	8.03E-15	0.04	4.52E-02	7.32	0.19	9.04	-5.581	-3.948	-14.245
Michigan2	5.740	0.020	8.92E-17	0.12	1.36E-01	7.84	10.00	8.87	-5.259	-4.398	-16.172
Michigan3	9.790	0.085	3.81E-16	0.01	1.58E-02	8.49	0.12	7.92	-4.786	-4.130	-15.522
Michigan4	12.930	0.097	4.35E-16	0.50	5.75E-01	8.49	0.15	6.73	-4.792	-4.012	-15.468
Alaska1	2.130	0.120	5.39E-16	1.00	1.13E+00	6.95	0.30	3.5	-6.555	-4.758	-15.335
Alaska2	0.160	0.029	1.30E-16	1.00	1.13E+00	7.28	0.20	5.0	-6.358	-5.861	-15.954
Alaska3	1.730	0.004	1.79E-17	1.00	1.13E+00	7.15	0.20	4.5	-6.329	-4.849	-16.830
Alaska4	0.330	0.069	3.10E-16	1.00	1.13E+00	7.91	0.20	9.5	-4.092	-5.683	-15.683
Alaska5	0.100	0.013	5.83E-17	1.00	1.13E+00	7.70	0.10	5.0	-5.515	-6.230	-16.451
Alaska6	1.010	0.005	2.24E-17	1.00	1.13E+00	6.99	6.00	4.0	-6.744	-5.060	-16.717
Illinois1	6.425	0.336	1.50E-15	0.04	4.52E-02	7.90	0.18	13.3	-4.785	-4.421	-14.994
Illinois2	25.180	0.272	1.22E-15	0.04	4.52E-02	7.44	0.18	14.2	-5.217	-3.839	-15.091
Massachusetts1	11.570	0.305	1.37E-15	1.00	1.13E+00	5.30	2.90	16.2	-8.732	-4.049	-14.972
Massachusetts2	20.340	0.071	3.18E-16	1.00	1.13E+00	5.70	9.10	13.3	-8.672	-3.784	-15.584
Massachusetts3	9.680	0.066	2.96E-16	0.02	2.94E-02	6.00	1.70	10.1	-7.950	-4.112	-15.624

TABLE A2. OUTPUT FILE FROM AQUEOUS GEOCHEMICAL MODELING SOFTWARE PROGRAM EQ 3/6 (WOLERY, 1992)

Name	Ra^{2+} Activity (pCi)	Ra^{2+} Activity (Bq)	Ra^{2+} (atoms/L)	Ra^{2+} (mol/L)	U^{4+} (mg/L)	UO_2^{2+} (mg/L)	NO_2^- (mg/L)	NO_3^- (mg/L)
Brazil	n/a	n/a	n/a	2.80E-15	8.35E-05	9.44E-05	n/a	n/a
Idaho 2	0.1400	0.005179430	378060603.10	6.28E-16	0.003110	0.0035143	0.010	0.947592
Idaho 4	0.2330	0.008620052	629200860.90	1.05E-15	0.001147	1.30E-03	n/a	n/a
Idaho 5	0.0360	0.001331853	97215583.66	1.61E-16	0.003977	4.49E-03	n/a	n/a
Idaho 6	0.0770	0.002848687	207933331.70	3.45E-16	1.300000	1.47E+00	0.010	0.17712
Idaho 7	-0.0005	-0.000018500	-1350216.44	-2.24E-18	1.000000	1.13E+00	0.010	0.95202
Idaho8	0.0130	0.000480947	35105627.43	5.83E-17	1.000000	1.13E+00	0.010	0.86346
Michigan1	1.7910	0.066259711	4836475287	8.03E-15	0.040000	4.52E-02	0.002	0.25682
Michigan2	0.0200	0.000739919	54008657.59	8.97E-17	0.120000	1.36E-01	0.008	3.63538
Michigan3	0.0850	0.003144654	229536794.70	3.81E-16	0.014000	1.58E-02	0.008	0.23025
Michigan4	0.0970	0.003588605	261941989.30	4.35E-16	0.509000	5.75E-01	0.004	0.24796
Alaska1	0.1200	0.004439512	324051945.50	5.38E-16	1.000000	1.13E+00	0.010	0.49150
Alaska2	0.0290	0.001072882	78312553.50	1.30E-16	1.000000	1.13E+00	0.010	0.17712
Alaska3	0.0040	0.000147984	10801731.52	1.79E-17	1.000000	1.13E+00	0.010	0.17712
Alaska4	0.0690	0.002552719	186329868.70	3.10E-16	1.000000	1.13E+00	0.010	0.17712
Alaska5	0.0130	0.000480947	35105627.43	5.83E-17	1.000000	1.13E+00	0.010	0.17712
Alaska6	0.0050	0.000184980	13502164.40	2.24E-17	1.000000	1.13E+00	0.010	0.28339
Illinois1	0.3360	0.012430633	907345447.50	1.51E-15	0.040000	4.52E-02	0.002	0.25682
Illinois2	0.2720	0.010062893	734517743.20	1.22E-15	0.040000	4.52E-02	0.002	0.25682
Massachusetts1	0.3050	0.011283759	823632028.20	1.37E-15	1.000000	1.13E+00	0.010	48.08365
Massachusetts2	0.0710	0.002626711	191730734.40	3.18E-16	1.000000	1.13E+00	0.010	36.73026
Massachusetts3	0.0660	0.002441731	178228570.00	2.96E-16	0.026000	2.94E-02	0.006	0.633204

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