

# Stochastic Environmental modeling for Nuclear Waste Management

by

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## **AUTHOR'S DECLARATION**

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

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## Abstract

Deep geological repositories are identified as possible disposal site for safely isolating highly radioactive nuclear waste from affecting humans and the environment. These repositories are multi barrier systems and safety of the system is very crucial since failure of the system will lead to radioactive contamination, which is harmful to the environment.

It is necessary to model the possible failure of the system, one of the most significant parameter is the mass transfer between the barriers in the multiple barrier system given by equivalent flow rates, half time of the solute and the delay time between the inflow and outflow of the barriers. The entire model is constructed based on the conservation assumption of mass flux. The model is used to analyze radioactive decays of the two long lived radioactive species C-14 (neutral non-sorbing nuclide) and I-129 (anionic non-sorbing nuclide). From the radioactive decay of these radionuclides the equivalent exposure is calculated to ensure that it is well below the current safety limits specified by the Regulator.

The geosphere and bentonite buffer, which are a part of the multi barrier system, are porous media and modeling the seepage is done using Darcy's law. Modeling seepage of water is important because water acts as a carrier for several elements that can potentially corrode the copper coating. The copper coating is an integral part of the multi barrier system, and an essential element of the used fuel container.

This thesis analyzes effects of a wide spectrum of uncertainties on the performance of the analytical solution obtained from the deterministic model is used to (i) consider parameter uncertainties, and (ii) derive stochastic solution of governing equations for the following two cases: (1) water seepage into the DGR, and (2) Mass outflow of radioactive material. Case I a man-made system whose uncertain and time invariant parameters, whereas Case II considers stochastic nature of the natural environment.

Conclusions from this study support a high level of safety aspects of DGR for the disposal of high level radioactive waste.

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## Nomenclature

<b>Symbol</b>	<b>Description</b>
$\lambda_c$	Decay constant from canister to buffer
$\lambda_{bt}$	Decay constant from buffer to tunnel
$\lambda_{bf}$	Decay constant from buffer to fracture
$\lambda_{tf}$	Decay constant from tunnel to fracture
$\lambda_f$	Decay constant from fracture to geosphere
$\delta_{t_{dc}}$	Delay time in canister
$\delta_{t_{abt}}$	Delay time from buffer to tunnel
$\delta_{t_{abf}}$	Delay time from buffer to fracture
$\delta_{t_{dtf}}$	Delay time from tunnel to fracture
$\delta_{t_{df}}$	Delay time from fracture to geosphere
$m_c$	Mass of radioactive material in canister
$m_b$	Mass of radioactive material in buffer
$m_t$	Mass of radioactive material in tunnel
$m_f$	Mass of radioactive material in fracture
$Q$	Volumetric flow rate
$k$	Permeability of porous medium
$A$	Cross section area of porous medium
$\mu$	Fluid viscosity
$p$	Pressure
$x$	Distance
$\varphi$	Porosity

$c_t$	Total compressibility
$c_f$	Compressibility of fluid
$c_\phi$	Compressibility of porous medium
$W_t$	Wiener process

## List of abbreviation

<b>Acronym</b>	<b>Description</b>
DGR	Deep Geological Repository
NWMO	Nuclear Waste Management Organization
HLW	High-Level Radioactive Waste
LLW	Low-Level Radioactive Waste
UFC	Used Fuel Container
IAEA	International Atomic Energy Agency
FEA	Finite Element Analysis
OPG	Ontario Power Generation
MIC	Microbe Induced Corrosion
APM	Adaptive Phase Management
HCB	Highly Compacted Bentonite
SRB	Sulfate Reducing Bacteria
ODE	Ordinary Differential Equation
PDE	Partial Differential Equation
MPM	Mixed Potential Model
GDSA	Generic Disposal System Analysis
DoE	Department of Energy
ICRP	International Commission on Radiological Protection

# Chapter 1

## Introduction

Electric power is generated in power stations, which produce electricity from turbines driven or fueled by combustion of coal, natural gas, nuclear fission, water energy (hydropower), wind and petroleum to name a few. Oil, coal and natural gas together form the highest fuel source for generating electricity around the world, they are also major contributors to carbon emission. With the growing concern of climate change, greenhouse effect and depleting natural resources, there has been an increase in electricity generation from nuclear power plants. In 1973 only 1.3% of the electricity produced in the world was from nuclear power plants, currently over 10% of the world's electricity is produced from nuclear power plants.

Nuclear power generation is getting very popular owing to its low carbon emission value, compared to coal and natural gas. There are currently 449 nuclear reactors in operation located in 31 countries for generating electricity and 60 reactors under construction. With the growing production of electricity using nuclear power generation, there is also the increase in radioactive waste which is the byproduct of nuclear power generation. Radioactive materials are harmful to humans and the environment, a material is classified as radioactive when it has unstable nuclei. This unstable nucleus decays to become stable, this is also known as radioactive decay. There are three types of decay, they are 1. Alpha decay where the radioactive nucleus emits alpha particles to become stable, 2. Beta decay which happens by positron emission or electron capture and 3. Spontaneous fission where heavier radioactive isotopes decay into lighter elements and two or three neutrons *Friedlander et al (1981)*. Radioactive waste are broadly classified into low level radioactive waste (LLW) and High Level Radioactive Waste (HLW). LLW includes filters, reactor water residue, protective garments, equipment and tools which are exposed to radioactive substances and become radioactive and these LLW do not have half-lives more than 5 years, hence the isolation and disposal is easier. The HLW consist of highly radioactive substances, which includes the used or spent nuclear fuel bundles and highly radioactive sludge which is obtained after reprocessing the used nuclear fuel bundles. A nuclear reactor which produces 1000 MW of electricity generates 27 tons of unprocessed HLW. They contain several short lived and long-lived fission products, among which Technetium-99 and Iodine-128 will become the major contributor for radioactivity after a few thousand years, since they have the highest half-lives- 220,00 years and 15.7 million years respectively. Currently most of the used nuclear fuel are stored in the reactor facility. The used fuel bundles release a lot of heat because of

radioactive decay, known as decay heat. Hence, they are stored in a pool of water for several years (between 7 to 10 years) to reduce the heat, after which they are stored in a dry condition in huge concrete tanks with steel covering. But these tanks only have a lifespan of 20 to 30 years. Long term isolation and containment of the radioactive used fuel bundles has been a growing concern for the past 4 to 5 decades. Several disposal methods have been researched by nuclear nations. One such method is long term storage above the ground, which is currently being done in most of the nuclear nations. Disposing the HLW into space, but it is expensive and highly risky in case of a failure during launch. Deep borehole disposal is a method of disposal similar to a DGR but the depth of the placement of the HLW is much greater than a DGR. Ocean or seabed disposal which was done by a few European nations but after the London convention of 1972, which was on prevention of marine pollution, it can no longer be done. Disposal in ice sheets was also considered for isolating HLW, but based on the Antarctic treaty, it is illegal to do so. One of the safest option is to place them in deep geological repositories (DGR), this has been accepted by several nuclear waste management agencies around the world. Low level radioactive waste has been isolated from the biosphere in DGR already in several nations. A DGR is being constructed in Finland by POSIVA which is Finland's nuclear waste management agency for isolating high-level radioactive waste. Other countries such as Canada, Japan, Sweden and the USA are planning to build DGRs for isolating high-level radioactive waste. The DGR is a multi barrier system, typically 350 to 500 meters below the ground with placement rooms or tunnels which have steel canisters coated with copper containing the used fuel bundles and the tunnels are sealed with a clay buffer, usually bentonite. The basic design concept is the above description, but it varies from country to country. For instance, the DGR in Finland uses a Mark I canister whereas the proposed Canadian DGR uses a Mark II canister. These DGR system is intended to contain the radioactive waste for a million years. The objective of this thesis is to model the effects of radioactive contamination on the environment in case the DGR fails.

The failure of the systems may occur 1. because of external factors such as groundwater entering the system which leads to corrosion of canister and causing a radioactive leakage or 2. because of radioactive leakage caused by inherent manufacturing defects. In this thesis, both cases are modeled, the first case is modeling groundwater affecting the system. Since the DGR is intended to last for over one million years, there are several factors which may change in the that long duration. Pressure on the system will change due to glacial pressure in case of an ice age, ground water parameters might change due to elevated temperature near the system and so on. In the second case, we assume the worst-case scenario of the canister to break causing a radioactive leakage. This is

modeled by using publicly available data for DGRs. Radioactive decay is considered a stochastic process because when several trials of counts recorded per second from a Geiger counter for long-lived radioactive isotope, there is no uniformity in the results. Which shows that the decay of a particular nuclei is completely a random event and statistical methods must be applied to fairly predict the time of decay for a given mass of radioactive material *Friedlander et al (1981)*, *Loveland et al (2005)*. Hence to make a realistic model these uncertainties should be incorporated. In simulation and modeling, first a deterministic model is developed then randomness is incorporated into it by considering the variance in different parameters. Stochastic modeling also helps in taking the uncertainties into account which we will see in detail in chapter 2 and chapter 3.

## **Chapter 2**

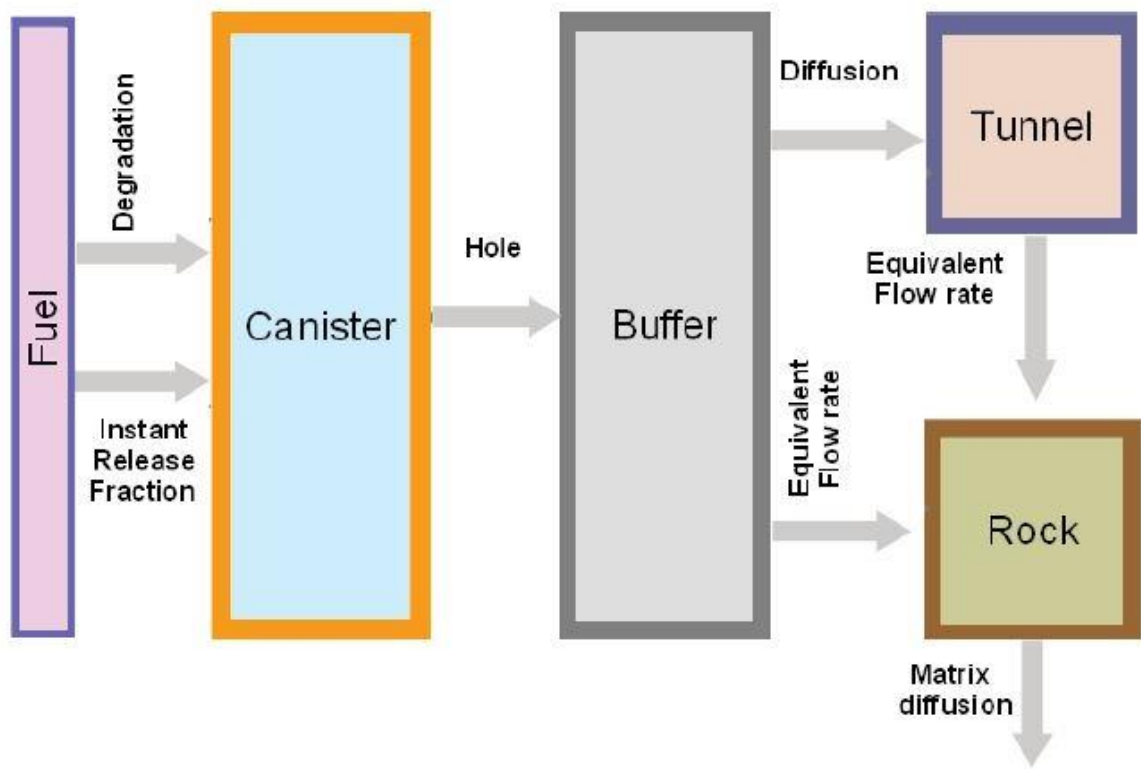
### **Literature review**

Every nation that produces power using nuclear plants has a plan to construct a deep geological repository to store and isolate the radioactive waste, which is the used fuel rods or the reprocessed waste. Some countries such as France, Russia, Japan, India and China reprocess the spent fuel to recover fissionable plutonium from the spent fuel. Whereas countries such as USA, Canada, Finland and Sweden do not reprocess the used fuel and plan to place them in the DGRs *Crowe et al (2013)*. Finland is the only nation which has started to build the DGR and will be placing the HLW starting in 2022, They have adopted Sweden's KBS-3 design for the multi barrier system. France and Sweden have decided on the site for the DGR. German had proposed a site but the project has been put on hold since 2011. But other nations are far from even having the design for their DGR approved or have a site for burial. In the United States, they were looking at the Yucca mountains in Nevada for constructing a DGR but it had a lot of opposition and finally the federal government stopped construction of the DGR in 2010 and is yet to reopen or find a new site (*Johnson, 2017*). Finding an appropriate site for a DGR is not only an engineering or geological problem but also a social problem. Canada's NWMO (Nuclear Waste Management Organization) has located 22 possible sites and it will be finalized only based on the approval of the local municipality, *NWMO (2015)*.

#### **2.1 Deep Geological repositories**

DGRs have been used for isolating LLW for over two decades. For HLW Finland is the first country to start construction of the DGR. POSIVA was established in 1995 in Finland for safe geological disposal of used nuclear fuel. The DGR is 350- 400 meters below the ground, it is based on SKB-3 a DGR design developed in Sweden. DGRs are multi-barrier systems which have one or two engineered barriers and also have the advantage of the geological surrounding, which acts as a third barrier for isolating radioactive waste for millions of years without the trouble of having to maintain the system.





**Fig 2.1 Flow Chart of multi-barrier system from POSIVA, Poteri et al (2012)**

From Fig 2.1 we can understand the multi barrier system and how each barrier interacts with one another. This is the basic design for a DGR which has been adopted in most of the countries. In countries which reprocess their used nuclear fuel, instead of the used fuel rods they will have highly radioactive slurry. The used nuclear fuel is placed in a canister which is then placed in a bentonite buffer which is surrounded by the rock matrix *Poteri et al (2012)*.

### **2.1.1 Fuel**

The high-level radioactive waste is the spent fuel from the nuclear reactor. As mentioned above, it is stored in cooling tanks for several years, followed by dry storage near the facility. In the DGR designs proposed by different countries, it is either reprocessed or placed directly in the canister. At the La Hague reprocessing plant in France, the high-level waste is calcinated and mixed with glass powder containing borosilicate and melted to temperatures over 1000°C. The molten mixture is then poured into steel canisters and stored away *Ojovan et al (2013)*. In Japan, they

reprocess the used fuel to extract plutonium by dissolving the fuel in nitric acid to obtain fissionable plutonium and high-level radioactive waste in liquid form, which is then vitrified before being placed in canisters *NUMO (2013)*. In Switzerland it is a similar vitrification process of liquid high-level waste after reprocessing *Nagra (2002)*.

In countries such as Finland and Canada where they do not reprocess the fuel, the used fuel rods are transferred to canisters that are filled with an inert gas such as argon and sealed. This process takes place above the ground in the proposed DGR's encapsulation plants *Posiva (2012)*. In the CANDU reactors in Canada, the fuel is in the form of pellets that are placed in long tubes called a fuel element made of Zircalory, which is an alloy of zirconium. The fuel element helps in isolating the used fuel pellets and 37 of these fuel elements are welded together to form a fuel bundle *NWMO (2011)*. This fuel bundle is then placed in the canister of the multi barrier system (Fig 2.2).

### **2.1.2 Canister**

The canister is the first or second barrier in the DGR depending on the used fuel, if it is reprocessed, the canister is the first barrier else, it is the second barrier. The canister design is very similar in most of the designs proposed by countries worldwide and have only a few design changes. The canister is an iron carbide container which has a copper coating for better corrosion resistance (*Keech et al 2014*). Evidences of natural copper found in the bedrocks without corroding has led to the use of a canister with copper coating. Copper is also known for its thermodynamic stability under DGR conditions. Posiva uses the Mark 1 canister which is made of nodular graphite cast iron insert which is strong enough to resist high mechanical stress caused by geological pressure inflicted by possible earthquakes or continental glacier. The use of cast iron also helps in the manufacturability of the canister.

In the Mark 1 design, a copper shell of thickness 5 cm facilitates as a leak-tight shell over the cast iron. A copper over pack having a lid and bottom is bolted to the shell to seal the canister *Posiva (2012)*. The canisters will be exposed to pressures as high as 44 MPa of which hydrostatic pressure and bentonite swelling pressure will contribute to 14 MPa. The canister will be exposed to an additional 30 MPa in case of an ice age causing ice sheets of thickness 3km. The mechanical property of this design has been studied and analyzed by several researchers. The probabilistic analysis on the cast iron insert shows that the probability of failure is in the order of  $2 * 10^{-9}$ , but collapse of the insert is strongly dependent on the assumed external pressure *Dillstrom (2005)*. The studies conducted *Raiko (2005)* shows that due to lack of material data, the performance of the inserts is

inconclusive when the given time scale is considered. Experimental results of creep test on the cast iron showed a maximum creep strain of 0.025% at a temperature of 125°C for a time span of 41,000 hr *Martinsson et al (2010)* but in real time conditions the cast iron will be exposed to temperatures as high as 70-80°C for several thousand years hence using Finite Element Method (FEM) for modeling the stress strain conditions will give a better understanding of the system. The creep failure of copper is more likely than that for cast iron, since copper is known for its low creep resistance *ASM (1990)*, *Avner (1964)*. Uniaxial copper creep deformation analysis using FEM computation for canisters has been done by *Sandstrom et al (2008)* which also includes multiaxial stress state which was further developed using novel approaches by *Jin et al (2009)*, *Raiko et al (2010)*. The results of the three models yield the same strain rate value, the order of  $10^{-12} \text{ s}^{-1}$ . The main drawbacks in the Mark 1 design is the enormous size of the canister, sharp edges in the end of the cylinder and bolting of the copper over pack in the top and bottom.

In the Mark 2 canister which is adopted in Canada, the design of the canister overcomes the drawbacks which are in the Mark 1 canister. The size of the Mark 2 is smaller and the end covers are hemispherical and welded to the cylindrical shell instead of the flat lid used in the Mark 1 *Oy et al (2003)*. A steel shell of thickness 25 mm is used in the Mark 2 canister which is coated with 3 mm of copper. The 3 mm of copper is coated by electro-deposition process. The hemispherical part is welded to the cylinder by single pass hybrid-laser-arc-welding process and the welded region is machined and coated with copper using cold spray technique *Crowe et al (2013)*. This is done to ensure the integrity of the canister since the heat affected zone will be the weakest part of the canister that can give in to high stress or corrosion *Lavalin (2011)*. Copper corrosion studies have revealed that 1.27 mm of copper is sufficient for protection from corrosion. It is found that the estimated maximum uniform corrosion was 0.17 mm, 0.1 mm of corrosion due to under deposition *King (2005)* and microbial induced corrosion contributing to 1 mm of the coating thickness for a period of 1 million years *King (1996)*, *King et al (1997)*. With a factor of safety, NWMO have decided to have a copper coating for 3 mm. However, these studies were done with little data on actual conditions that may exist in the DGR.

### **2.1.3 Buffer**

Between the copper canister and the rock matrix, a buffer material is in place. The buffer is made of bentonite. Bentonite clay is used because of its desirable properties such as low hydraulic conductivity, high swelling ability, rheological stability, high plasticity and good thermal conductivity

*Appelo (2013)*. The compacted bentonite is expected to swell when in contact with water because of the clay-water interaction, this swelling is required for the sealing of the repository *Kanno et al (1999)*. The bentonite is considered to be highly compacted when it has a dry density greater than  $1500 \text{ Kg/m}^3$ , it is preferred for the buffer material since it is with low free water which brings down the water activity, preventing the growth of microbes *Pusch (1980)*. In the Posiva design, the compacted bentonite is used in the form of blocks while in the NWMO design, it canister is surrounded by a box of highly compacted bentonite. The drawback in using blocks of bentonite is, that it is time consuming and transporting all the blocks to the site will be an arduous process since it may swell up if it is not transported in dry conditions. In the buffer box encapsulation of the canister, uniformity of bentonite property can be achieved and transportation is easier. In the NWMO design (Fig 2.3) between two buffer boxes, a spacer box is placed for better heat dissipation in the repository. The walls of the placement room are planned to be lined with bentonite clay called the gap fill to ensure a more complete sealing of the placement room. One of the issues in the gap fill is that the dry density of the gap fill cannot be determined with certainty since it may vary along the height of the placement room and bentonite property changes significantly with respect to the dry density *Kaufhold (2013)*.

Modeling the behavior of bentonite is an interesting study, since highly compacted bentonite may have very low permeability but for the time scale of reliability that is needed in this system (1 million year) the bentonite is going to be saturated with water within 500 years *Pusch (1983)*. Case I in this thesis models the seepage of water into the DGR.

#### **2.1.4 Rock matrix**

The rock matrix is the surrounding bedrock of the DGR. Site selection for the DGR is one important step since the characteristics of the rock matrix will highly influence the DGR system. As mentioned before, the DGR is built 300 to 500 m below the surface. Soil sampling for geophysical investigation and groundwater investigation must be carried out extensively before deciding on the site. DGRs for LLW has been in use for the past two decades, the LLW DGR site in Germany and USA is in a salt dome. In Finland, the LLW is stored in granite (Crystalline) bedrock. While in Canada for LLW, OPG DGR site is in a limestone bedrock. For HLW, only a couple of countries have decided on the site, Finland which has started the construction of the DGR in Olkiluoto the rock matrix is granite. Similarly, Sweden has proposed to construct the DGR in a granite bedrock. In Canada, the site could either be sedimentary or crystalline.

Sedimentary rocks are more saline compared to crystalline rocks, high salinity in the bedrock will prevent microbial activity *Manger et al (1963)* in water there by protecting the copper from MIC. The crystalline rocks have the desirable property of high thermal conductivity and crystalline rocks are also very dry compared to sedimentary rocks *Nasir et al (2014)*.

### 2.1.5 Fluid flow through porous media

Groundwater flow is very similar to fluid flowing through a porous media. It can be modelled by using Darcy's law which is applicable for creep or Stokes flows which have Reynolds number less than one *Tyrkko (2009)*. Darcy's law is given by Equation 2.1. The law is based on some assumptions which are

1. Aquifer material is incompressible
2. Water is incompressible
3. External load is a constant
4. It is a non-leaky aquifer
5. Hydraulic conductivity is isotropic
6. Porous media is already saturated

$$Q = \frac{-KA \partial p}{\mu \partial x} \quad \text{Equation 2.1}$$

This can be further represented as a partial differential equation by incorporating the concept of mass conservation, which gives equation 2.2

$$\frac{\partial p}{\partial x} = \frac{K}{\phi \mu c_t} \frac{\partial^2 p}{\partial x^2} \quad \text{Equation 2.2}$$

Where

Q = Volumetric flow rate

k = Permeability of porous medium

A = Cross Section area of porous medium

$\mu$  = Fluid viscosity

p = Pressure

x = Distance

$\phi$  = Porosity

$c_t = c_f + c_\phi$   $c_f$  = Compressibility of fluid,  $c_\phi$  = Compressibility of porous media

The volumetric flow rate is the desired parameter which is to be calculated. Permeability is the ability of a medium to let fluid flow through it, it is represented as 'k' and the SI unit is  $m^2$ . The permeability of a given material varies depending on porosity, stress, temperature and the fluid which is flowing through it. Bentonite's permeability depends on its dry density *Villar M V et al (2005)*. The bentonite having a dry density of  $1410 \text{ kg/m}^3$  has a permeability of  $5 \cdot 10^{-12} \text{ m}^2$  and that with a higher dry density of  $1700 \text{ kg/m}^3$  has a lower permeability of  $4 \cdot 10^{-12} \text{ m}^2$  *Eloranta (2012)*.

Viscosity is the magnitude of internal friction in the fluid, fluid with high viscosity have a greater resistance to flow. It is represented as ' $\mu$ ' and the SI unit is Pa.s. Viscosity of a fluid depends on the temperature and pressure in the surrounding. Water has a viscosity of  $8.90 \cdot 10^{-4} \text{ Pa.s}$  at  $25^\circ \text{C}$  and it decreases with increase in temperature.

Porosity is the ratio of void volume to material volume in a solid. In the case of bentonite, highly compacted bentonite has lower porosity, bentonite with a dry density of  $1410 \text{ kg/m}^3$  has porosity of in the range of 45-50% and that with a dry density of  $1700 \text{ kg/m}^3$  has a porosity of 35-40% *Kaufhold S et al (2013)*.

Compressibility is the measure of change in volume of a material when pressure is applied on it. It can be represented as the inverse of bulk modulus. Compressibility of water at  $25^\circ \text{C}$  is  $45.8 \cdot 10^{-11} \text{ Pa}^{-1}$

### **2.1.6 Mathematical model for mass outflow in DGR**

Researchers have been studying about this multi-barrier system since the 1980's. *Nilsson et al. (1991)* is one of the earliest works on the SKB-3 design, where the model is designed to study the steady state transport of the radionuclide from a single defective or corroded canister through the buffer, fracture and into the rock matrix. The model compares the transportation of the nuclide through the barriers to a resistance network model hence simplifying a complex 3-dimensional

numerical model. POSIVA modeled the system by taking into consideration the mass transfer between the barriers of the system. The species' considered to represent and understand radioactive decay in this model are C14-non sorbing nuclide, I129- anion and dominating nuclide, Pu239 sorbing nuclide. The numerical model is a chain radioactive decay, since each of the engineered barrier can be represented in terms of the half-life of the solute, mass transfer capability of the barrier and the delay time. Considering the delay time while modeling makes it more realistic since it takes time for the solute to reach the outflow location, it is a time shift in the release rate of the solute from the barrier. The basic decay chain reaction is expressed by the following equation *Poteri (2012)*.

$$\begin{aligned}
 \frac{dm_c}{dt} + \lambda_c m_c &= \delta_0 \\
 \frac{dm_b}{dt} + (\lambda_{bf} + \lambda_{bt}) m_b &= \lambda_c m_c * \delta_{t_{dc}} \\
 \frac{dm_t}{dt} + \lambda_{tf} m_t &= \lambda_{bt} m_b * \delta_{t_{dbt}} \\
 \frac{dm_f}{dt} + \lambda_f m_f &= \lambda_{tf} m_t * \delta_{t_{dtf}} + \lambda_{bf} m_b * \delta_{t_{dbf}}
 \end{aligned}
 \tag{Equation 2.3}$$

Where

- $\lambda$  = Decay constant of the solute
- $m$  = Mass transfer coefficient
- $*$  = Convolution operation
- $\delta_t$  = Dirac delta function  $\delta_\tau = \delta(t-\tau)$
- $c$  = Canister
- $b$  = Buffer
- $t$  = Tunnel
- $f$  = Fracture or Geosphere

The solution for chain ODE in equation 2.3 is given in equation 2.4 where  $m_{out}$  is the final mass outflow through all the barriers, which is nothing but  $m_f$  in equation 2.3.

$$\begin{aligned}
m_{out} = & H(t - t_{d1})\lambda_c\lambda_{bt}\lambda_{tf}\lambda_f \left[ \frac{e^{-(t-t_{d1})\lambda_c}}{(\lambda_{bt} - \lambda_c)(\lambda_{tf} - \lambda_c)(\lambda_f - \lambda_c)} \right. \\
& + \frac{e^{-(t-t_{d1})\lambda_{bt}}}{(\lambda_c - \lambda_{bt})(\lambda_{tf} - \lambda_{bt})(\lambda_f - \lambda_{bt})} \\
& + \frac{e^{-(t-t_{d1})\lambda_{tf}}}{(\lambda_c - \lambda_{tf})(\lambda_{bt} - \lambda_{tf})(\lambda_f - \lambda_{tf})} \\
& \left. + \frac{e^{-(t-t_{d1})\lambda_f}}{(\lambda_c - \lambda_f)(\lambda_{bt} - \lambda_f)(\lambda_{tf} - \lambda_f)} \right] \\
& + H(t - t_{d2})\lambda_c\lambda_{bf}\lambda_f \left[ \frac{e^{-(t-t_{d2})\lambda_c}}{(\lambda_{bf} - \lambda_c)(\lambda_f - \lambda_c)} \right. \\
& \left. + \frac{e^{-(t-t_{d2})\lambda_{bf}}}{(\lambda_c - \lambda_{bf})(\lambda_f - \lambda_{bf})} + \frac{e^{-(t-t_{d2})\lambda_f}}{(\lambda_c - \lambda_f)(\lambda_{bf} - \lambda_f)} \right]
\end{aligned}$$

Equation 2.4

$$t_{d1} = t_{dc} + t_{abt} + t_{atf} + t_{af}$$

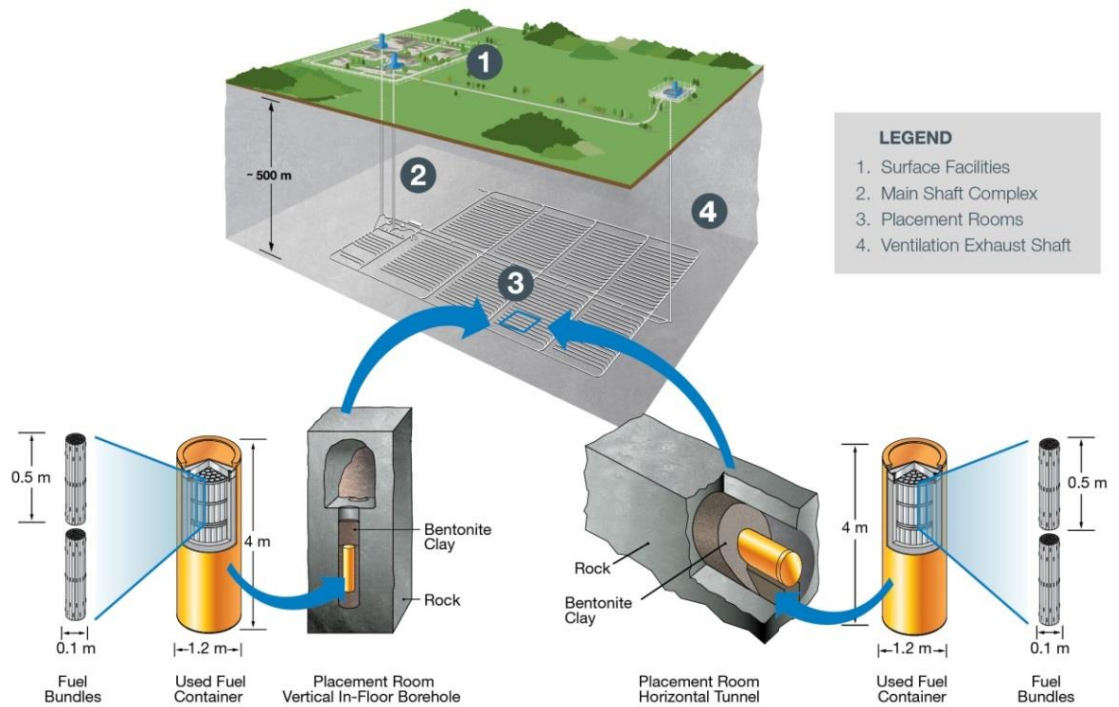
$$t_{d2} = t_{dc} + t_{abf} + t_{af}$$

Where H is the Heaviside step function and the delay time is given by each  $t_d$  represents the delay time between each of the barriers.

The model from POSIVA is only a deterministic model and does not account to any sort of uncertainty in the system. In the following chapter, we will see in detail how the same base model behaved when uncertainty was incorporated in it.

The Canadian DGR design is different from the Finnish or Swedish design, however, the general model form doesn't change as it contains a similar barrier system. NWMO's Adaptive Phased Management (APM) plan for used nuclear fuel was accepted by the Canadian federal government in 2007. APM plan focusses on the technical methods and management system of the repository. As per the proposed design by NWMO, the repository is to be built 500 meters below the ground, in the Canadian shield, they have not decided on the exact location for building the repository yet.

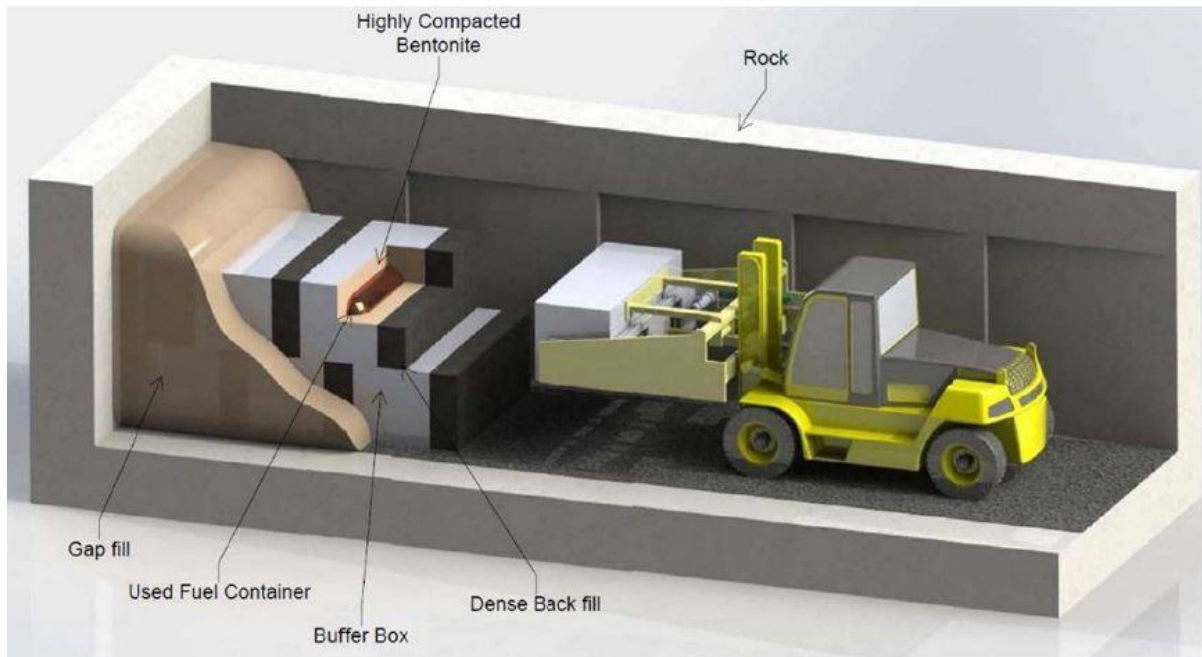




**Fig 2.2 Canadian Deep Geological Repository (Source: NWMO)**

The Fig 2.2 illustrates the design of the repository, the fuel bundles are natural uranium made into pellets and packed in the form of a bundle. They are very safe before being loaded into the reactor and can be handled without any special equipment. But they become radioactive after they are taken out of the reactor. As of mid-2016, Canada has an inventory of 2.7 million used fuel bundles and produce 90,000 bundles each year. A steel canister holds 48 of such used fuel bundles. The canisters are coated with 3-millimeter copper coating by cold spray technique. Copper is chosen for the coating of the steel canister because it is one of the naturally occurring metal, which is found in stable condition under the ground for several years. The canister is then placed in a buffer box made of highly compacted bentonite (HCB). The bentonite also helps to slow down water transport to the canister as it has desirable properties such as low permeability and sealing capability as it swells up when in contact with water *Harrington et al (2003)*. Each buffer box is separated by a spacer box, which helps in heat dissipation. The placement room is then sealed and closed with a gap fill material made of bentonite of lesser dry density (Fig 2.3). Finally, it is the rock matrix which surrounds the entire system. Similar to the SKB-3 design, the Canadian repository is also a multi barrier system.

The only difference is in the design of the system, such as the canister design and use of buffer box instead of block of bentonite as buffer. Since a site has not been selected yet, the type of rock which will surround the repository is unknown. It could be either crystalline or sedimentary and each has its own advantage and disadvantage to the entire system. Crystalline rocks have higher thermal conductivity compared to sedimentary rocks. Sedimentary rocks are more saline compared to crystalline rocks, and high salinity is a preferred parameter as it prevents microbial activity in groundwater. Presence of sulfate reducing bacteria (SRB) is one of the critical parameter since it leads to corrosion of the copper coating. Groundwater will be main carrier of these microbes and other chemical agents which can corrode the copper *King et al (2007) (2001)*.



**Fig 2.3 Placement room**

## 2.2 Radiation units

There are several quantities represented in different units related to radiation. Absorbed dose is the mean energy imparted to certain mass of substance by the ionizing radiation. Absorbed dose is essential to be calculated since it relates radiation and its effect on any substance. Radiation protection is defined by the International Atomic Energy Agency as “The protection of people from harmful effects of exposure to ionizing radiation, and the means for achieving this”. The exposure to radiation can be limited based on the duration of exposure, distance from the source and amount of shielding. Effective dose can be reduced by decreasing the time of exposure, increasing the distance from the source or having a thick shielding. Given below are some of the commonly used quantities to represent radiation. *Quantities, (1971)*.

### 2.2.1 Activity

The term activity, which is also known as specific activity, is the activity of a radionuclide hence it is the physical property of an isotope, the SI unit for activity is Becquerel (Bq) which is the reciprocal of seconds. Becquerel is the number of radioactive transformation that occurs per second. The non-SI unit to represent activity is Curie (Ci). It is defined as the  $1 \text{ Ci} = 3.7 * 10^{10}$  decay per second. Hence  $1 \text{ Ci} = 3.7 * 10^{10} \text{ Bq}$ . Similar to Curie, Rutherford (Rd) is another non-SI unit which is defined as the activity of a quantity of radioactive material in which one million nuclei decay per second.

### 2.2.2 Fluence

It is amount of energy reserved by a surface per unit area from a radiation source. Fluence is denoted by “ $\phi$ ” and the unit for fluence is reciprocal of area ( $\text{m}^{-2}$ )

### 2.2.3 Absorbed dosage

It is the amount of energy absorbed by one unit of mass. The SI unit for absorbed dose (D) is gray (Gy) which is defined as the absorption of “one joule of radiation energy per kilogram of matter”. The non-SI unit for absorbed dosage is Rad and  $1 \text{ rad} = 0.01 \text{ Gy}$ , it is defined in CGS system as “the dose causing 100 ergs of energy to be absorbed by one gram of matter”

### 2.2.4 Exposure

Exposure is defined as the ionization of air due to ionizing radiation from a radioactive material. It is represented in the unit Roentgen (R ) and is defined as “the quantity of radiation which

liberates by ionization of one electrostatic unit of charge per  $\text{cm}^2$  of air under normal temperature and pressure. This unit for representing radiation is not very popular, since it depends on the radiated particle such as alpha, beta or gamma. It is approximately,  $1 \text{ R} = 10 \text{ mSv}$

### 2.2.5 Dosage equivalent

All the above quantities where either with respect to a specific kind of radiated particle or radiation absorbed by objects of certain mass. Dosage equivalent (H) gives effect of ionizing radiation on the human body. Since it considers the biological effectiveness of the radiation it is widely used for representing allowable safety limits. The SI unit for representing dosage equivalent is Sievert (Sv), It is the probability of the risk of cancer or genetic damage that can be inflicted by exposure to radiation. The SI base unit for Sievert is  $\text{m}^2 \text{ s}^{-1}$ . The CGS unit to represent dose equivalent is Rontgen equivalent man (rem).  $1 \text{ Sv} = 100 \text{ rem}$ .

## 2.3 Finite difference method

Various physical phenomena such as heat, fluid dynamics, sound, electrostatics and quantum mechanics can be mathematically represented as partial differential equations (PDE) similar to equation 2.1 and 2.2. They are multi variable functions having partial derivatives, it represents rate of change of a continuous variable. Unlike ordinary differential equation (ODE) where the unknown depends on a single independent variable, in PDE the unknown function depends on two or more independent variables hence solving PDEs are more challenging. Finite difference method is one such numerical method to find the approximate solution of PDEs, other such numerical methods are finite element method and finite volume method. For solving equation 2.2, we will be using finite difference method in the following chapters.

Finite difference method uses finite difference to find the approximate solution to differential equations. That is the partial differentiation of  $y$  over  $x$  can be represented as  $\frac{\partial y}{\partial x} = \frac{y_{i+1} - y_i}{\Delta x}$

$$\text{Similarly, } \frac{\partial^2 y}{\partial x^2} = \frac{y_{i+1}^n - 2y_i^n + y_{i-1}^n}{\Delta x^2} .$$

Where “i” represents the node and “n” represents time step. Finite difference method can be classified into two types, 1. Explicit method and 2. Implicit method. The explicit method uses forward difference and the implicit method uses backward difference.

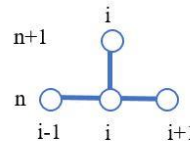
### 2.3.1 Explicit method

Let us use the example of the following PDE, which is a one-dimensional parabolic PDE.

$$\frac{\partial y}{\partial t} = \frac{\partial^2 y}{\partial x^2} \quad \text{Equation 2.5}$$

Assuming the initial condition to be  $y(x,0) = f(x)$  and boundary conditions to be  $y(0, t) = y(1, t) = 0$ .

$$\frac{y_i^{n+1} - y_i^n}{\Delta t} = \frac{y_{i+1}^n - 2y_i^n + y_{i-1}^n}{\Delta x^2}$$



Equation 2.6

The equation evaluates  $y_i^{n+1}$  for all nodes given in  $y_i^n$ . In the explicit method, the equations can be written in the form of a vector matrix to solve the PDE.

$$y^{n+1} = A Y^n$$

Where A is  $\frac{\Delta t}{\Delta x^2}$

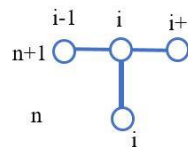
$$\begin{bmatrix} -2 & 1 & 0 & 0 & 0 \\ 1 & -2 & 1 & 0 & 0 \\ 0 & 1 & -2 & 1 & 0 \\ & & & 1 & -2 & 1 \\ & & & 0 & 1 & -2 \end{bmatrix}$$

Where  $Y^n$  is the matrix with the  $y$  values at time state  $n$  and node  $i$ . The initial condition and boundary conditions must be selected based on the physical problem to ensure that the errors will be minimum.

### 2.3.2 Implicit method

Using the same example, when we use backward difference in time and central difference in space, we will get the below equation. Where the superscript represents time and the subscript represents the node.

$$\frac{y_i^{n+1} - y_i^n}{\Delta t} = \frac{y_{i+1}^{n+1} - 2y_i^{n+1} + y_{i-1}^{n+1}}{\Delta x^2}$$



Equation 2.7

The above equation is solved to get  $y_i^{n+1}$  for the given  $y_i^n$  since we used backward deference to expand the PDE. The linear system is solved at every time step and the implicit method is more numerically stable than the explicit method *Hoffman et al (2001)*

## 2.4 Modeling of DGR

Deterministic models help as a base model to study and understand the system and for helping in simulation for several thousand years, such as the work conducted by *Ahn (2007)* which gives the environmental impacts of the Yucca mountain DGR in terms of radiotoxicity using a deterministic model. *Lin et al (2007)* modeled a DGR system for analyzing the reliability of system when oxygen diffuses through the barriers to the canister, they also suggested the use of multi physics modeling software such as COMSOL for modeling such system. One of the drawbacks in using such commercial software is that solving a probabilistic model or a stochastic model takes a significant CPU time.

### 2.4.1 Uncertainty

In modelling, when uncertainties are taken into consideration the outcome of the model is more realistic. There could be several factors which could contribute to the uncertainty in the model. Uncertainty is classified based on its origin as parameter uncertainty, model inadequacy, residual variability, parametric variability, observation error and code uncertainty by *Kennedy et al (2001)*. In the context of modeling, uncertainty is usually classified as aleatory uncertainty and epistemic uncertainty. Aleatory uncertainty is caused by inherent randomness in the system and can't be reduced. Epistemic uncertainty is caused because of lack of knowledge or unavailability of data and it can be reduced by improving the physical model, calibration etc. by *Drzewiecki (2013)*. Aleatory uncertainty can be modeled by considering a random solution model which can assume a basic random variable to the input variable. In DGR development, different agencies from around the world have listed the various uncertainties which may affect the system performance *Nagra (2002)*. The Mixed Potential Model (MPM) developed by DoE, USA goes only to the extent of sensitivity analysis on the electrochemical reactions on the surface of the canister (*Wang et al 2014*). The Generic Disposal System Analysis (GDSA) developed on PLOTRAN by the DoE allows the analysis of the UFC in various rock matrix like salt bed and clay (*Marnier et al. 2015*)

The work done by *Helton et al (2012)* gives stochastic results of performance analysis of the Yucca mountain repository in the USA by listing various failure scenarios and running the model for each of the scenarios. Stochastic modeling can be done for considering the epistemic uncertainty in the system. In this thesis, we will be seeing both aleatory and epistemic uncertainty in the DGR system. *Helton et al (2014)* also worked on probabilistic models to predict the failure of the DGR system for various failure scenarios.

## 2.4.2 Stochastic modeling

When modeling a physical system by incorporating uncertainties in them, the model must be described by means of a stochastic differential equation which converts a deterministic differential equation such as equation 2.8, in to 2.9 in which  $X_t$  is the stochastic variable.

Stochastic modeling helps in incorporating uncertainty in physical systems. If equation 2.8 represents a physical system, by introducing a stochastic process  $N_t$ , we can model the initially deterministic model as a stochastic model as given in equation 2.9

$$\begin{aligned} \frac{dx}{dt} &= f(x_t, t) && \text{Equation 2.8} \\ x(t_0) &= x_0 \end{aligned}$$

$$\begin{aligned} \frac{dX}{dt} &= f(X_t, t) + g(X_t, t)N_t && \text{Equation 2.9} \\ X_{t_0} &= X_0 \end{aligned}$$

In equation 2.9, the initial condition  $X_0$  can also be a random variable.

If the function  $f(x_t, t)$  represents a physical system where  $x$  is the process,  $K$  is a parameter and  $s$  is the source, the stochastic differential equation can be written as equation 2.10 if the uncertainty is associated with the source.

$$\frac{dX_t}{dt} = K X_t + s_t + \sigma N_t \quad \text{Equation 2.10}$$

$$X_{t_0} = X_0$$

In equation 2.10 the uncertainty is introduced by adding white noise ( $N_t$ ) with some intensity ( $\sigma$ ) to the source  $s_t$ .

Similarly, if the uncertainty is to be introduced to the parameter, white noise is added to the parameter  $K$  (Equation 2.11)

$$\frac{dX_t}{dt} = (K + \sigma N_t) X_t + s_t \quad \text{Equation 2.11}$$

$$X_{t_0} = X_0$$

In the following chapter, we will be discussing about Wiener process or standard Brownian motion in section 3.3.2, where we introduce a Wiener process  $W_t$  which has a stationary independent increment  $W_t - W_{t-1}$  which is a Gaussian random variable. Consider the initial physical system in equation 2.8 to represent it as a stochastic process it can be written as equation 2.12 where  $g(X_t, t)$  is function which specifies the noise and  $\partial\beta_t$  is the Brownian motion. Where  $\partial\beta_t = W_t dt$ .

$$dX_t = f(X_t, t)dt + g(X_t, t) W_t dt \quad \text{Equation 2.12}$$

Equation 2.12 can be solved by stochastic integration between  $0 \leq t \leq \infty$  to obtain  $X_t$

$$X_t = X_0 + \int_{t_0}^t f(X_s, s)ds + \int_{t_0}^t g(X_s, s)\partial\beta_t \quad \text{Equation 2.13}$$

In equation 2.13,  $\int_{t_0}^t g(X_s, s)W_s$  is stochastic integration whereas the previous part is regular integration, equation 2.14 gives the solution for the stochastic integral. Two of the well-known ways of solving stochastic integrals is Ito and Stratonovich.

$$\int_{t_0}^t g(X_s, s)\partial\beta_t = \lim_{\Delta t \rightarrow 0} \sum_{i=0}^{n-1} g(x, t'_i) (\beta_{t_{i+1}} - \beta_{t_i}) \quad \text{Equation 2.14}$$

The difference between Ito and Stratonovich is that in Ito the evaluation point of the integral is chosen in the beginning of the interval  $(t_0, t)$  where as in Stratonovich the evaluation point is chosen in the



middle of the interval at  $t_{\frac{1}{2}} = \frac{1}{2}(t_0, t)$ . It is easier to make formal and theoretical calculations using Ito and Stratonovich connects well with the rules of differentiation and integration of ODE *Stijnen, et al (2003)*.

Since a DGR is a very complicated system, which needs a multi-disciplinary analysis, to ensure the reliability of the system, in this thesis Case II considers a large coefficient of variation (standard deviation over mean) to analyze and study the DGR system. Though several researches have worked on the hydro-mechanical-chemical perspective of the engineered barriers, this thesis gives the results of a detailed epistemic uncertainty analysis for water seepage into the DGR. In Chapter 3 we will see how the model for case I and case II are developed and Chapter 4 gives the results of these models.

## Chapter 3

### Model Development

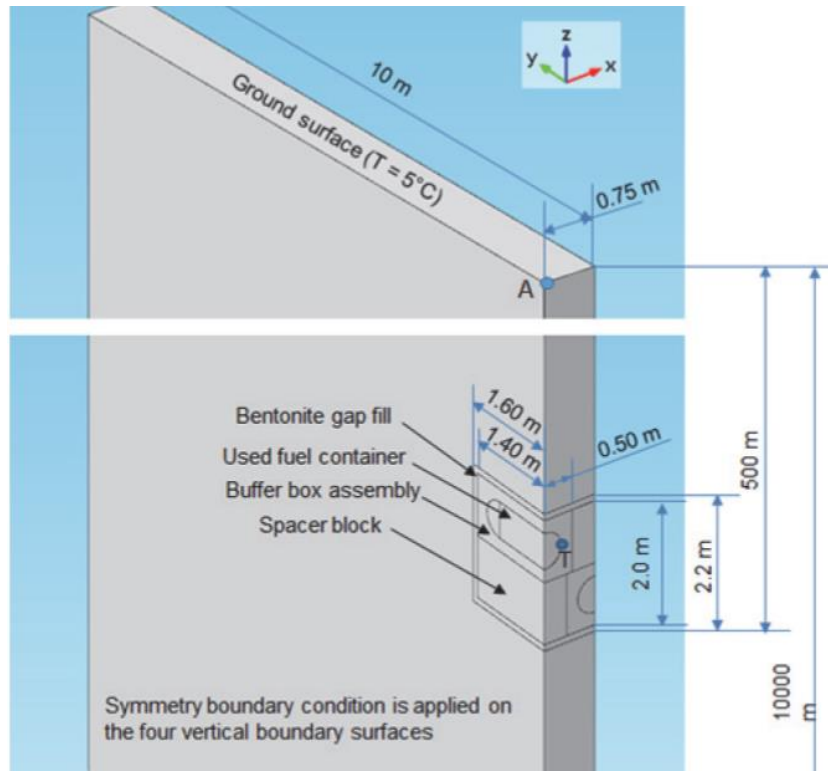
In this chapter, we will see how the model was developed for Case I which is modeling water seepage into the system using Darcy's law and for Case II which is the worst-case design, where we use Equation 2.4 to model the mass transfer between the barriers, assuming that the canister breaks as soon as the DGR is closed and for both the cases, we made a base model which is deterministic, followed by a random model and then a stochastic model

#### 3.1 Deterministic model

It is essential to make a base model which is deterministic while modeling uncertainty. For both the cases we used appropriate deterministic models. For Case I, the base model was Darcy's law given in Equation 2.2. For Case II the base model was the mass transfer chain reaction from Equation 2.4.

##### 3.1.1 Case I: Modeling water Seepage into the DGR

For modeling water entering the DGR system, we will use Darcy's law in equation 2.2. Since ground water flow through rocks is like fluid flow through porous medium. From literature, we have acquired the necessary data for using Darcy's law (given in Table 3.1). A finite difference method of the implicit type is applied to Equation 2.2. For the model, we assume an initial pressure to be 1 atmosphere. On the right-hand side, the boundary condition in the model is given such that the gradient is zero so that the model is continuous. The time domain is in the day units and the model simulates for 10 years with a  $\partial t$  of 0.5. The length domain given in meters for a length of 0.15m with  $\partial x$  of  $5 \times 10^{-4}$ . We take such small values of  $\partial t$  and  $\partial x$  so that the model is more accurate. The results of the model are presented in section 4.1



**Fig 3.1 Detailed Drawing of Repository**

**Table 3.1 Parameters for modeling water seepage**

Parameter	Units	Gap fill	Buffer box
Dry density	Kg/m <sup>3</sup>	1410	1700
Permeability K	m <sup>2</sup>	5*10 <sup>-12</sup>	4*10 <sup>-12</sup>
Porosity		0.3613	0.2943
Viscosity $\mu$ (H <sub>2</sub> O)	mPa	1.002 to 0.7978 at 20-30°C	
Compressibility $c_t$	Pa <sup>-1</sup>	48.84 X 10 <sup>-11</sup>	48.03 X 10 <sup>-11</sup>

### 3.1.2 Case II: Modeling mass outflow of radioactive material from DGR to Geosphere

From the literature, we obtain the decay chain reaction through the engineered barrier and the values of delay time and decay constant for two long lived radioactive nuclide which would be the major contributors to radioactivity in the used fuel bundles. The two-radioactive nuclide considered in the model are C-14 and I-129. The decay constant and delay time values are given in Table 3.1. Decay constants are proportional to equivalent flow rate and volume of the respective barrier. Delay time is proportional to the distance, retardation coefficient and diffusion coefficient. The deterministic model is developed by using the values from Table 3.2 in Equation 2.4.

**Table 3.2 Parameters for modeling mass outflow**

Parameter	Notation	Unit	C-14	I-129
Solute decay constant from canister to buffer	$\lambda_c = \frac{q_c}{V_c}$	1/Year	$3.8121 \times 10^{-4}$	$3.3663 \times 10^{-5}$
Solute decay constant from buffer to tunnel	$\lambda_{bt} = \frac{q_{bt}}{R_{bp}\epsilon_b V_{bt}}$		0.0273	0.0023
Solute decay constant from tunnel to fracture	$\lambda_{tf} = \frac{q_{tf}}{R_{pt}\epsilon_t V_t}$		41.8642	41.8642
Solute decay constant from fracture to geosphere	$\lambda_f = \frac{1}{4.3 u^2}$		0.0032	0.0316
Delay time from canister to buffer	$t_{dc}$	Year	$4.7089 \times 10^{-6}$	$4.7089 \times 10^{-6}$

Delay time from buffer to tunnel floor	$t_{dbt}$		0.1308	1.5696
Delay time from tunnel to fracture	$t_{dtf}$		0	0
Delay time in fracture	$t_{df}$		7.3567	0.7357

$q_c$ - Equivalent flow rate in canister

$V_{bt}$ - Volume of buffer

$V_c$ - Volume of canister

$q_{tf}$ - Equivalent flow rate from tunnel to fracture

$q_{bt}$ - Equivalent flow rate from buffer to tunnel

$R_{pt}$ - Retardation coefficient in tunnel

$R_{bp}$ - Retardation coefficient in buffer

$\varepsilon_t$ - Porosity of tunnel

$\varepsilon_b$ - Porosity of buffer

$V_t$ - Volume of tunnel

$u^2$  Transport resistance

The delay time from tunnel to the fracture is omitted and taken as zero, since any radioactive material that reaches the tunnel is going to affect the fracture based on the transport resistance ( $u^2$ ) hence delay time from tunnel to fracture is taken as zero and delay time in the fracture is alone considered in the last barrier.

## **3.2 Random Parameter model**

The DGR sites must be reliable for a million years and there are so many factors which contribute to the reliability of the system. Hence it is necessary to consider all the uncertainties which could result in the failure of the system. To incorporate uncertainty in the model, the parameters are given a variance and the deterministic model is converted into a random model. This is done for each parameter and all the parameters together so that we can understand the sensitivity of each parameter on the model.

### **3.2.1 Case I: Modeling water seepage into the DGR**

The parameters in equation 2.2 can vary with time because of pressure and temperature. From equation 2.1 we know that volumetric flow rate is proportional to viscosity and viscosity of water decreases with increase in temperature. We know that radioactive decay releases heat and the temperature in the placement room can go as high as 70-80°C. Geothermal heat is also a factor in elevated temperature in the DGR. The base model for water seepage is modified by multiplying the numerical values with a chosen coefficient of variation to make the random models. Sensitivity analysis is done to find the parameter which affects the system the most and section 4.1.2 gives the detailed results of the random model.

### **3.2.2 Case II: Modeling mass outflow of radioactive material**

Parameter in equation 2.4 may vary with time because of thermal, hydro or chemical factors. It can be incorporated in the deterministic model by multiplying the numerical values of decay constant between barriers and delay time between barriers with coefficient of variance. Sensitivity analysis is done to find out which parameter affects the system the most. Section 4.2.2 gives the detailed results of this model.

### 3.3 Stochastic model

#### 3.3.1 Case I: Modeling water seepage into the DGR

For case I, modeling water seepage into the DGR the uncertainty in the model is only epistemic hence stochastic modeling was not done for this case and the uncertainty will only affect the initial conditions such as the rock matrix, pressure, temperature, saturation etc.

#### 3.3.2 Case II: Modeling mass outflow of Radioactive material

Since Equation 2.4 is dependent on time, it can be written as Wiener process which makes the model stochastic. It is also called a standard Brownian motion (as we saw in section 2.4) replacing  $t$  in equation 2.4 with  $W_t$  we get equation 3.1 where  $W_t = \{ W_t, t > 0 \}$ ,  $W_0 = 0$  and the increment  $W_s - W_t$  is a Gaussian random variable with mean = 0 and variance =  $t-s$ . That is  $E [W_t - W_s] = 0$  and  $\text{var} [W_t - W_s] = t-s$ . The value of  $W_t$  changes at every time instance by a factor of the Gaussian random variable with mean zero and variance  $dt$ . In section 4.2.3 gives results from this model and derivative details can be found in Ponnambalam, et al. (2010).

$$\begin{aligned}
 m_{out} = & H(w_t - t_{d1}) \lambda_c \lambda_{bt} \lambda_{tf} \lambda_f \left[ \frac{e^{-(w_t - t_{d1}) \lambda_c}}{(\lambda_{bt} - \lambda_c)(\lambda_{tf} - \lambda_c)(\lambda_f - \lambda_c)} \right. \\
 & + \frac{e^{-(w_t - t_{d1}) \lambda_{bt}}}{(\lambda_c - \lambda_{bt})(\lambda_{tf} - \lambda_{bt})(\lambda_f - \lambda_{bt})} \\
 & + \frac{e^{-(w_t - t_{d1}) \lambda_{tf}}}{(\lambda_c - \lambda_{tf})(\lambda_{bt} - \lambda_{tf})(\lambda_f - \lambda_{tf})} \\
 & \left. + \frac{e^{-(w_t - t_{d1}) \lambda_f}}{(\lambda_c - \lambda_f)(\lambda_{bt} - \lambda_f)(\lambda_{tf} - \lambda_f)} \right] \\
 & + H(w_t - t_{d2}) \lambda_c \lambda_{bf} \lambda_f \left[ \frac{e^{-(w_t - t_{d2}) \lambda_c}}{(\lambda_{bf} - \lambda_c)(\lambda_f - \lambda_c)} \right. \\
 & \left. + \frac{e^{-(w_t - t_{d2}) \lambda_{bf}}}{(\lambda_c - \lambda_{bf})(\lambda_f - \lambda_{bf})} + \frac{e^{-(w_t - t_{d2}) \lambda_f}}{(\lambda_c - \lambda_f)(\lambda_{bf} - \lambda_f)} \right]
 \end{aligned}$$

Equation 3.1

## **Chapter 4**

### **Results and Discussions**

#### **4.1 Case I Results**

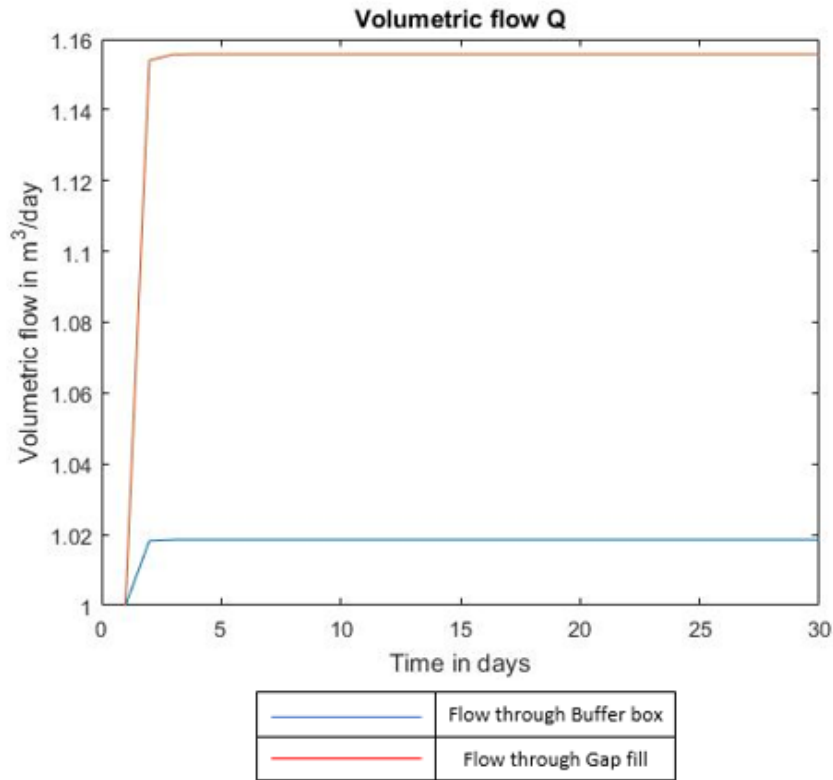
In this section, the results from Case I on water seepage into the DGR is discussed for the deterministic model and the random parameter model.

##### **4.1.1 Deterministic model**

The deterministic model is developed based on Darcy's law, which gives the value of volumetric flow rate. The results give one dimensional volumetric flow of water into the DGR through the gap fill material and the buffer box assuming initial pressure of 1 atmosphere. Fig 4.1 gives the steady state flow under saturated condition for gap fill of thickness 15 cm and buffer box of thickness 20cm.

From Table 3.1 we know that the bentonite used in the gap fill is of lower dry density compared to the bentonite in the buffer box. The two bentonite material were studied separately and it is evident from Fig 4.1 that flow through the buffer box is lesser compared to the gap fill. Since the gap fill surrounds the bentonite, the flow will originate from the gap fill and then to the buffer box (Fig 3.1). Though the flow in the gap fill is higher, since the downstream flow is lesser the flow in the gap fill will reduce to that in the buffer box.



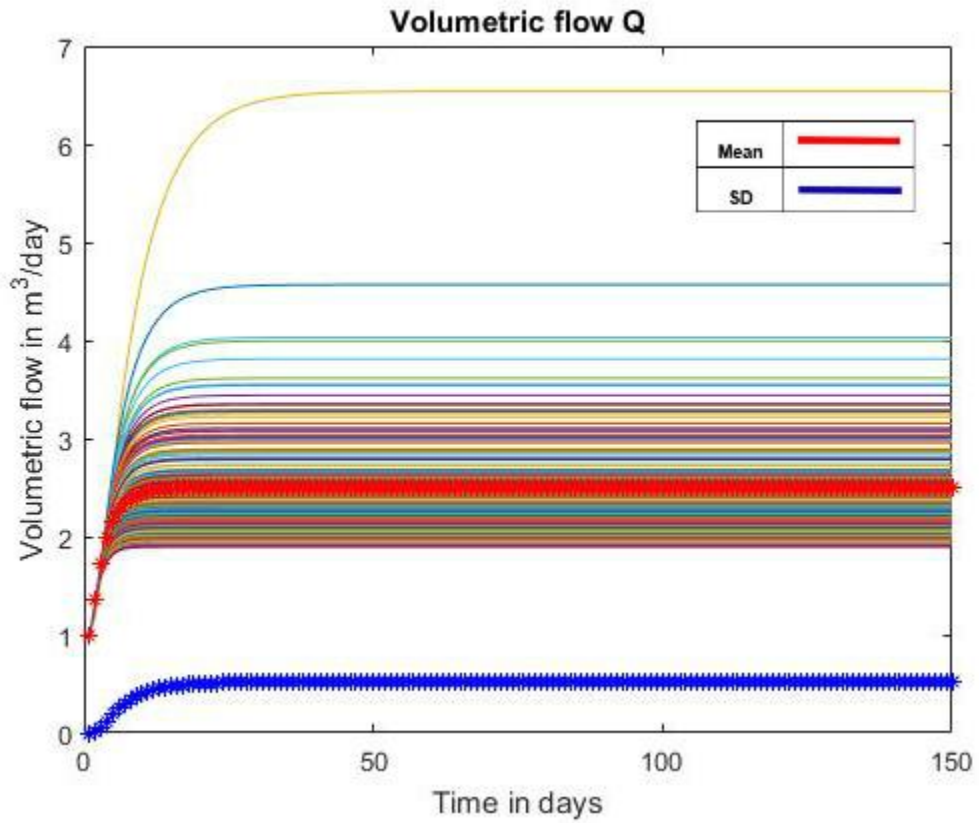


**Fig 4.1 Steady state flow under saturated condition**

#### 4.1.2 Random parameter model

From the Chapter 2 we know that the parameters such as permeability, viscosity, porosity and compressibility in Equation 2.1 and 2.2 are subjected to change based on other factors such as temperature and pressure. Under the ground, temperatures will be elevated because of geothermal gradient, which is about 25°C per km. That is, temperature below the surface increases by 25°C for every kilometer. Added to the geothermal gradient, temperature in the placement rooms will also be higher because of the decay heat generated from the canisters containing the used nuclear fuel but these are all to be considered uncertain. The random parameter model helps in considering these epistemic uncertainties.

The deterministic model was used as the base model and the parameters were incorporated with 30% variance using gaussian distribution. The results from 200 runs of the model is given in Fig 4.2



**Fig 4.2 Water seepage model for 200 runs with parameters having 30% variance**

**Table 4.1 Numerical results for Random parameter model**

Parameter	Volumetric flow	
	m <sup>3</sup> /day	m <sup>3</sup> /year
Mean	2.6955	983.8575
Max	7.2578	2649.097
Min	2.0105	733.8325
Standard deviation	0.5908	215.642

Assuming the placement room to be of the dimension 500\*4\*2.5 meters, it will be of the volume 5000 m<sup>3</sup>. The time it will take for one placement room to be flooded with water in case of a one-dimensional seepage from the rock matrix is given in Table 4.2. which shows that the average time it will take for the placement room to be flooded with water is 5 years.

**Table 4.2 Flooding of a placement room**

Parameter	Volumetric flow	Flooding of placement room
	m <sup>3</sup> /year	Years
Mean	983.8575	5.0820
Max	2649.097	1.8874
Min	733.8325	6.8135

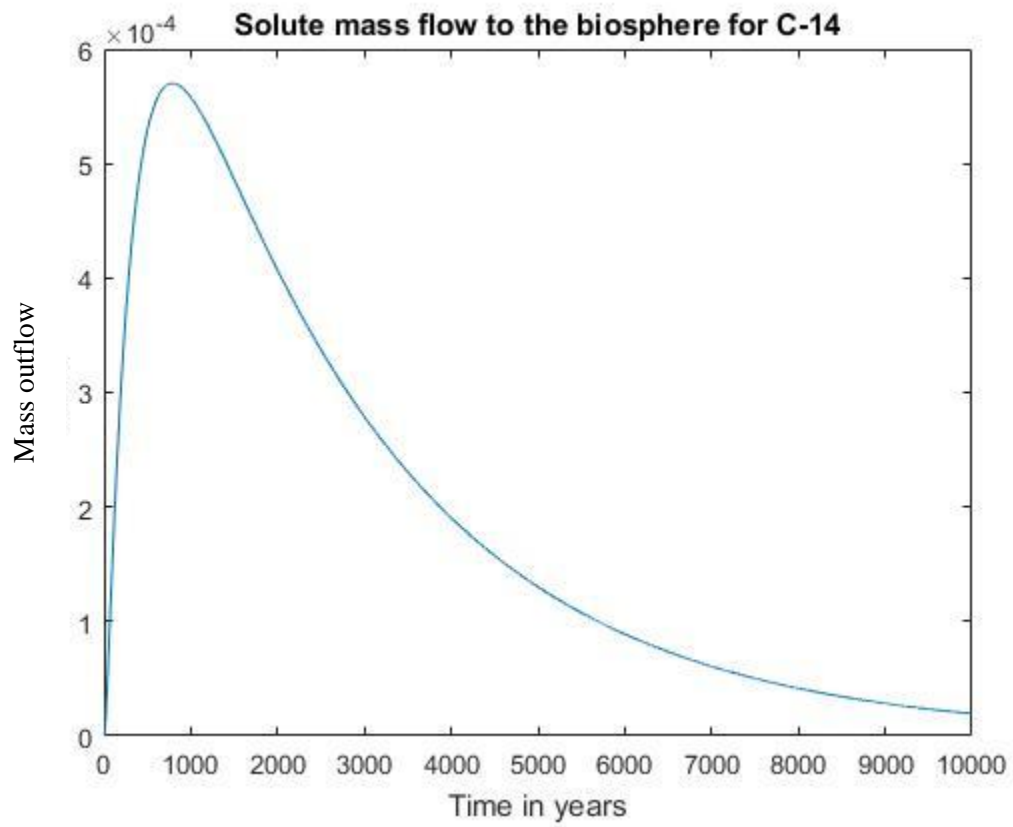
## 4.2 Case II results

This section comprises of all the results for Case II which is modeling the radioactive exposure if the canister fails as soon as the DGR is closed. The results for the deterministic model, random model and the stochastic model are given in the following sections.

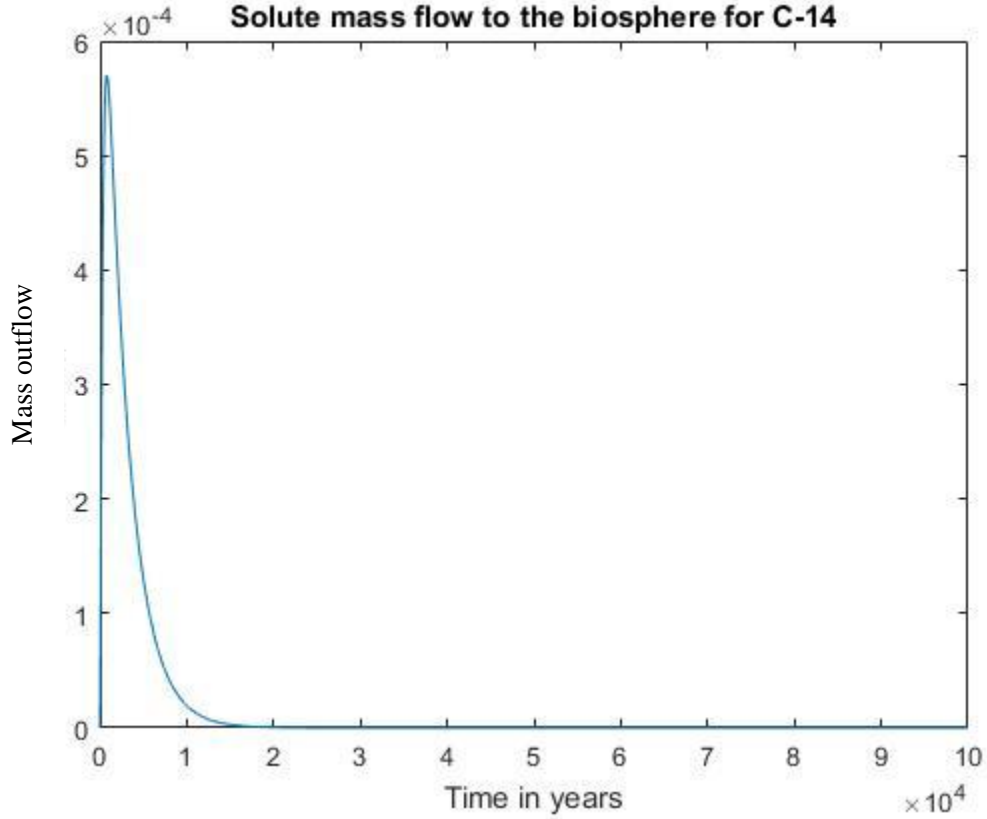
### 4.2.1 Deterministic Model

The deterministic model for the mass outflow of radioactive material is done based on Equation 2.4 using the data from Table 3.1 per canister. Later we can multiply these values for the entire system. The species considered are C-14 which is a non-sorbing nuclide and I-129 which is an anion and dominating nuclide. Equation 2.4 gives the mass outflow into the biosphere by taking the total response function considering chain decay first order ordinary differential equation. The response function is computed for the two considered species for 1 million years.

The response function in Fig 4.3 is the mass outflow of C-14 for the first 10000 years, assuming the systems fails and starts to leak radioactive material from the canister. Similarly, Fig 4.4 gives the mass outflow of C-14 in hundred thousand years. It is evident from the response curve that maximum outflow occurs at 789 years with an outflow value of  $5.7006 \times 10^{-4}$  1/year.

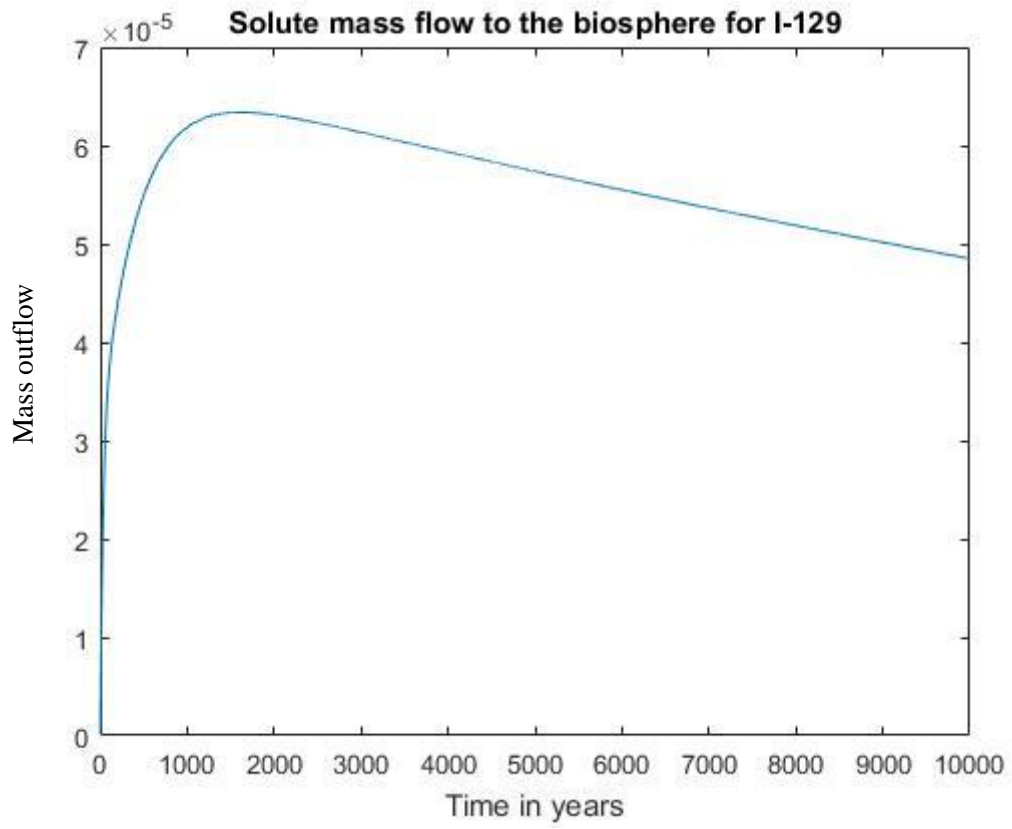


**Fig 4.3 Mass outflow of C-14 for the first 10000 years**

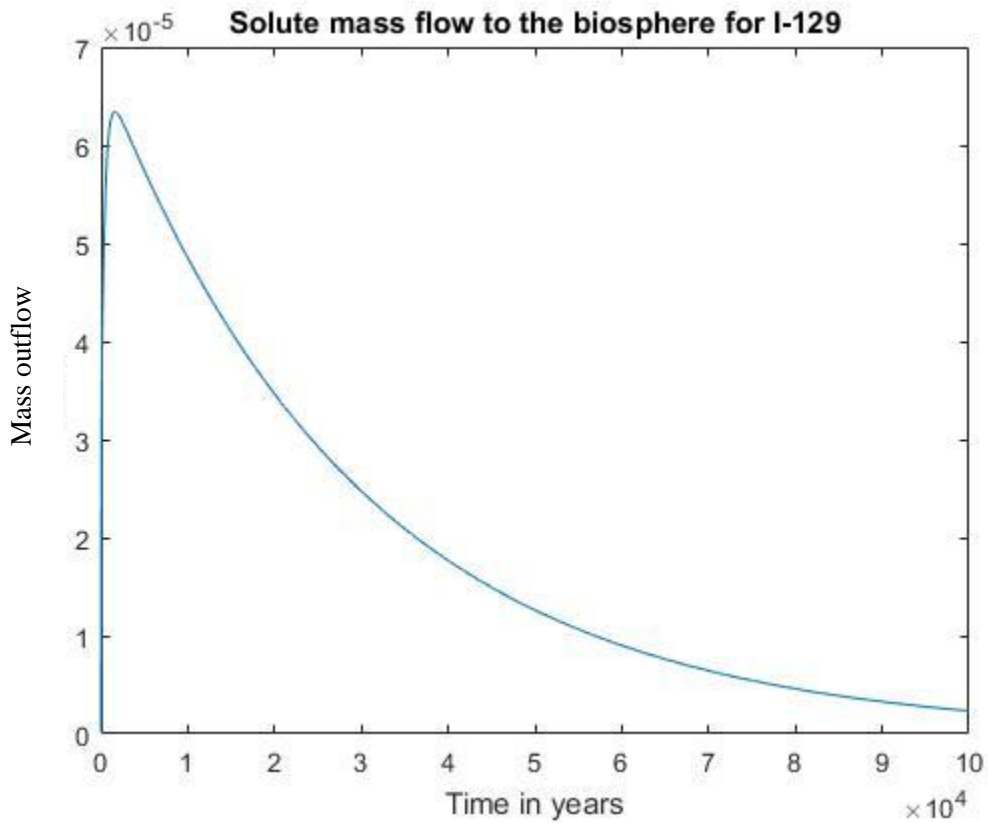


**Fig 4.4 Mass outflow of C-14 for hundred thousand years**

Fig 4.5 and Fig 4.6 shows the response curve for I-129 for 10000 years and hundred thousand years, respectively. Since the half-life of the considered species (5370 years for C-14 and 15.7 million years for I-129) are different, the response curves are different. Maximum outflow for I-129 occurs in 1608 years based on the deterministic model with an outflow of  $6.3378 \times 10^{-5}$  1/year.



**Fig 4.5 Mass outflow of I-129 for ten thousand years**

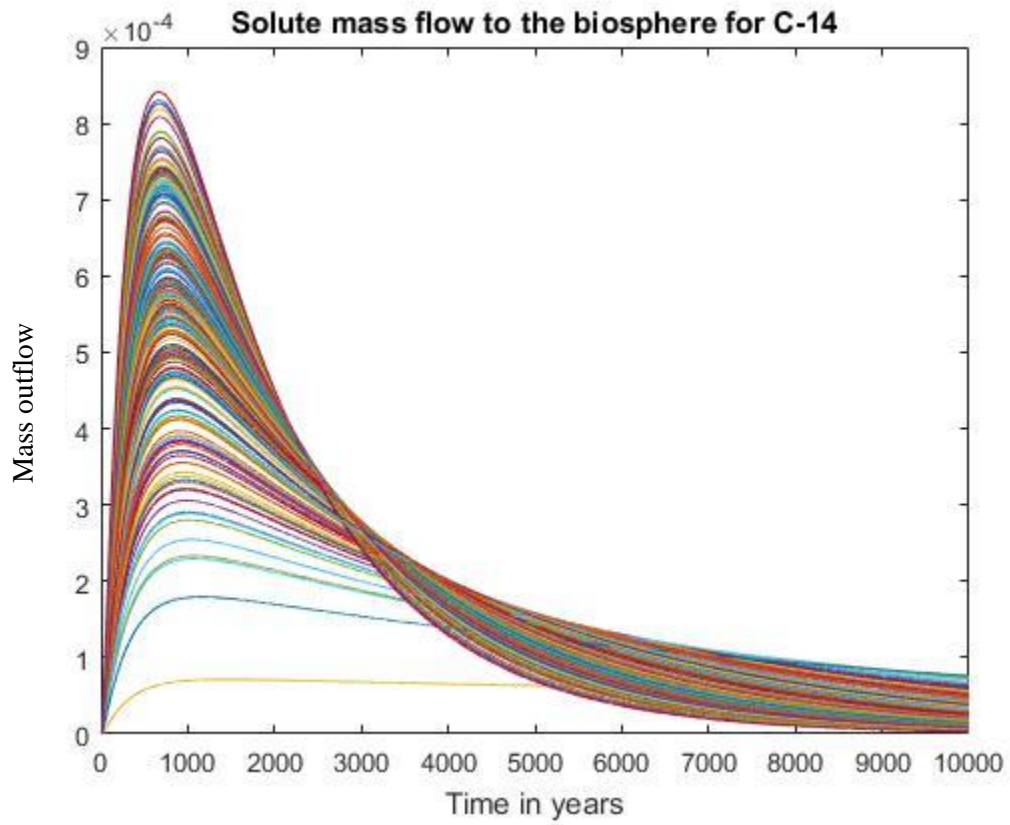


**Fig 4.6 Mass outflow of I-129 for hundred thousand years**

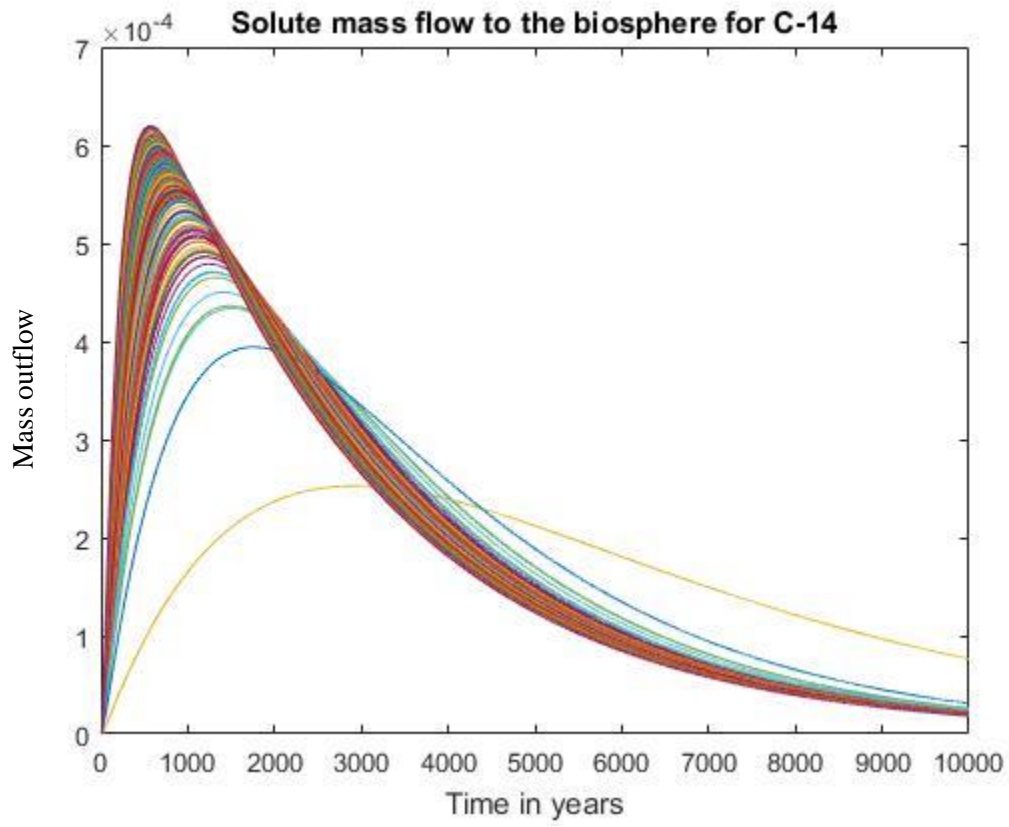
#### **4.2.2 Random Parameter model**

As explained in section 3.2.2 in the random model for case II, each parameter in Equation 2.4 is incorporated with a variance of 30% to study the behavior of the of the system with uncertainties. This also helps in identifying which parameter affects the system the most. Fig 4.7, Fig 4.8, Fig 4.9 and Fig 4.10 gives the response curve for C-14 when the mass outflow terms from each of the barriers are incorporated with a 30% variance.

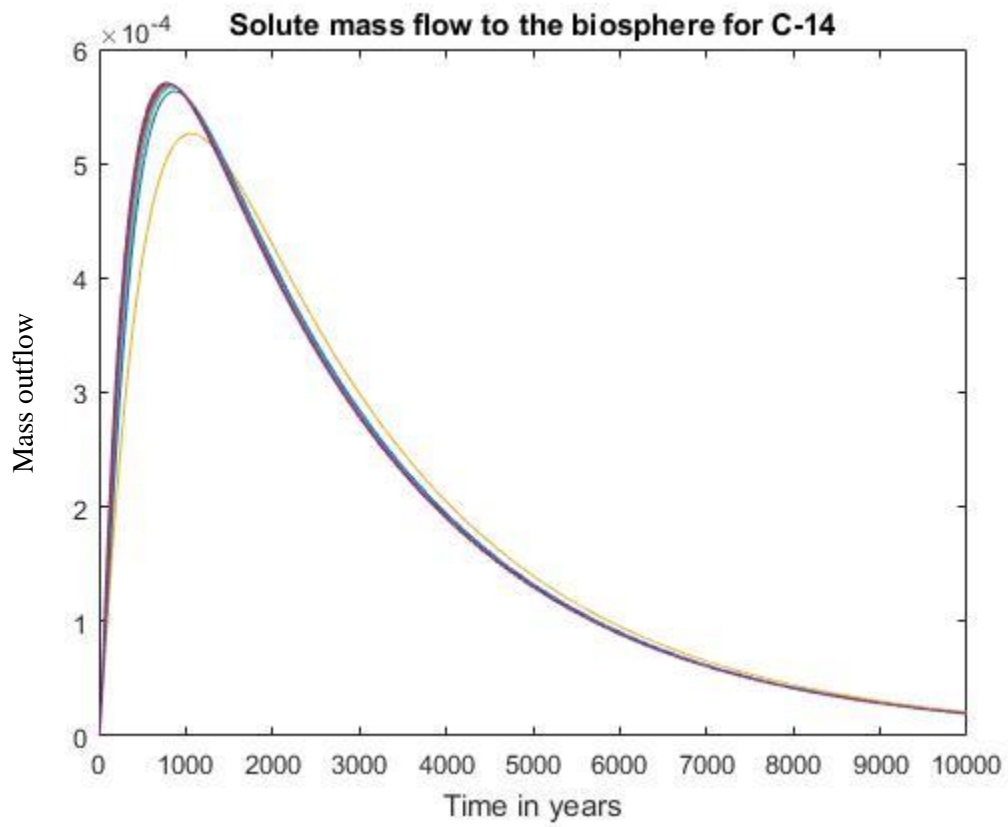




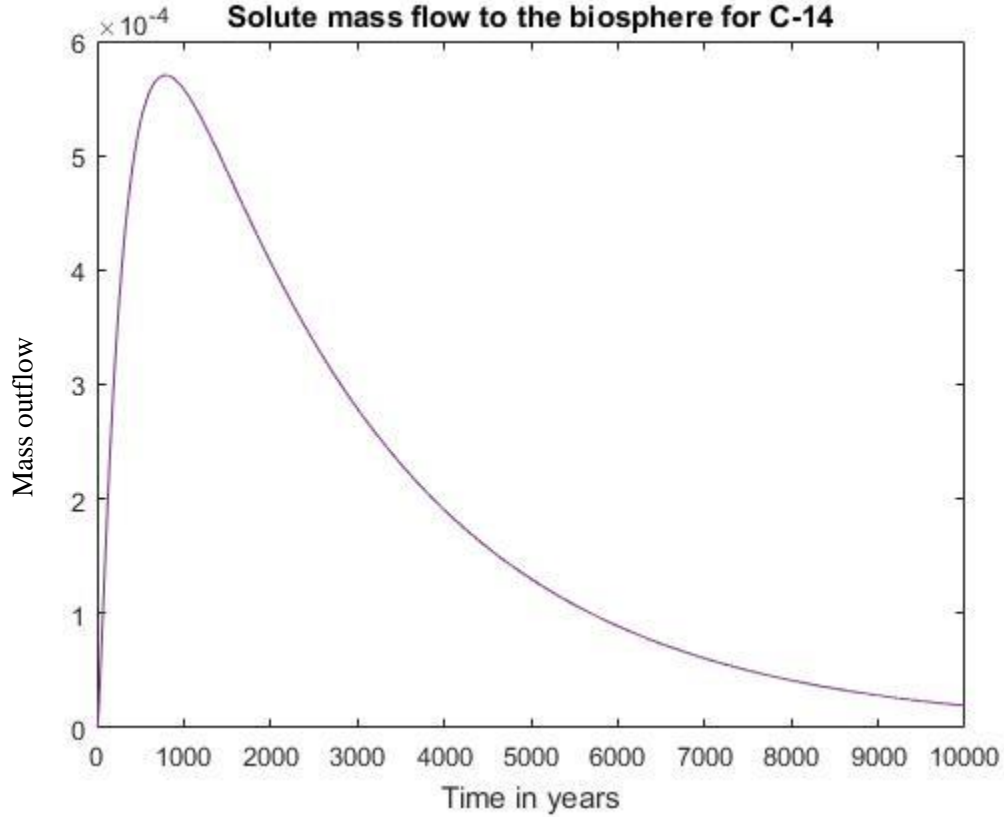
**Fig 4.7 Mass outflow with  $\lambda_c$  having variance of 30%**



**Fig 4.8 Mass outflow with  $\lambda_t$  having variance of 30%**



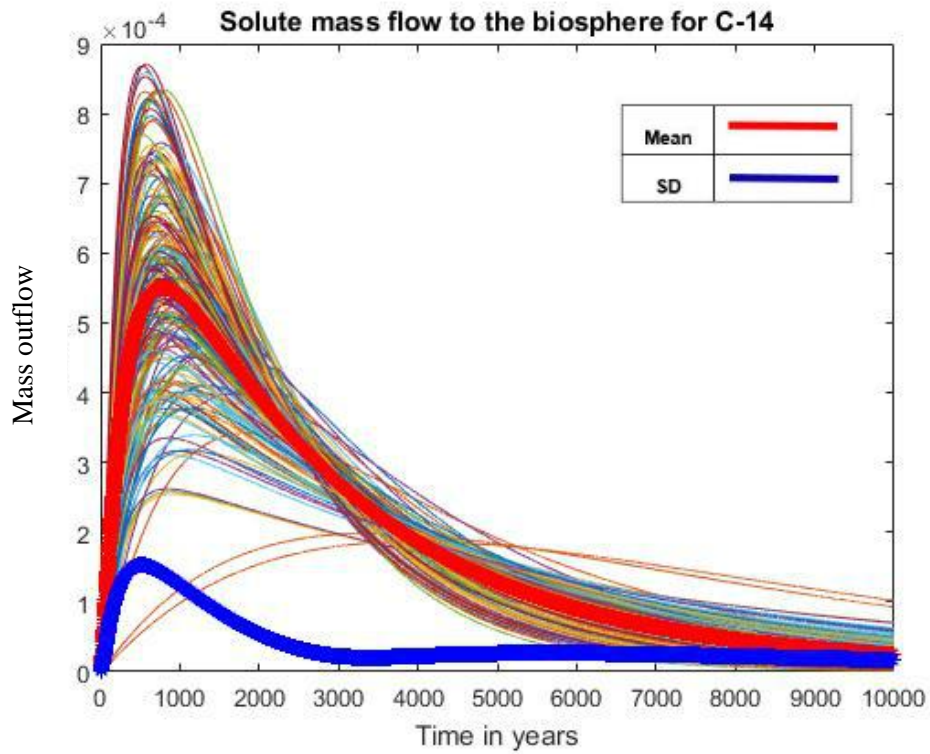
**Fig 4.9 Mass outflow with  $\lambda_{bt}$  having variance of 30%**



**Fig 4.10 Mass outflow with  $\lambda_{df}$  having variance of 30%**

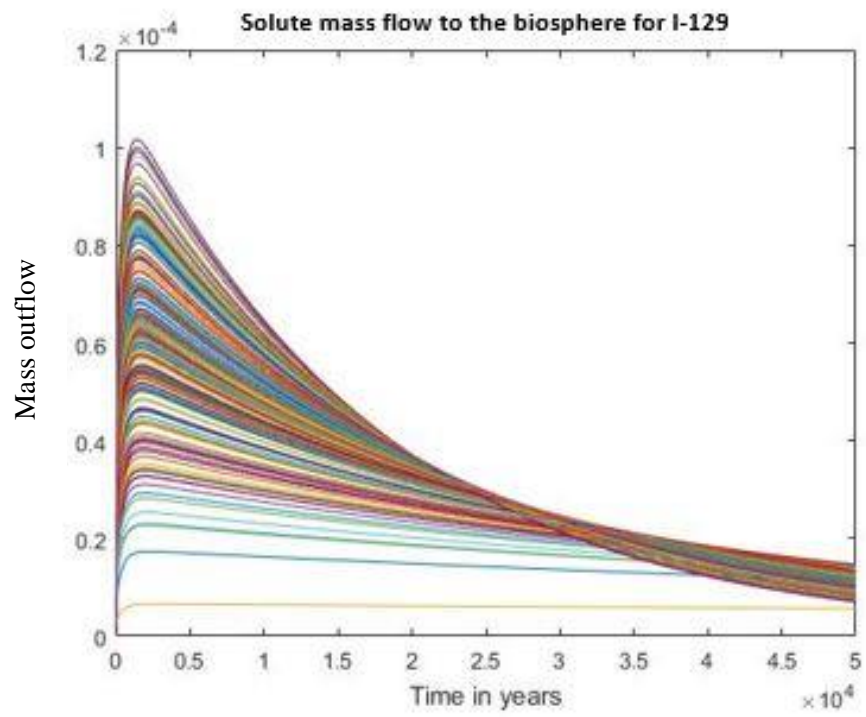
From the above mass outflow curves, it is evident that the parameter  $\lambda_c$  (Fig 4.7) is the most sensitive of all the parameters and the response curve is unaffected by any changes in parameter  $\lambda_{df}$  (Fig 4.10). The same applies to the delay time parameters, since they do not directly contribute to the mass outflow (See section 2.1.6).

Fig 4.11 give the response curve for C-14 for 200 runs having a variance of 30% in all the parameters, note that the model runs for ten thousand years, after which there was no significant outflow. The peak value of mass outflow from the 200 runs is  $8.7050 \times 10^{-4}$  at 576 years.

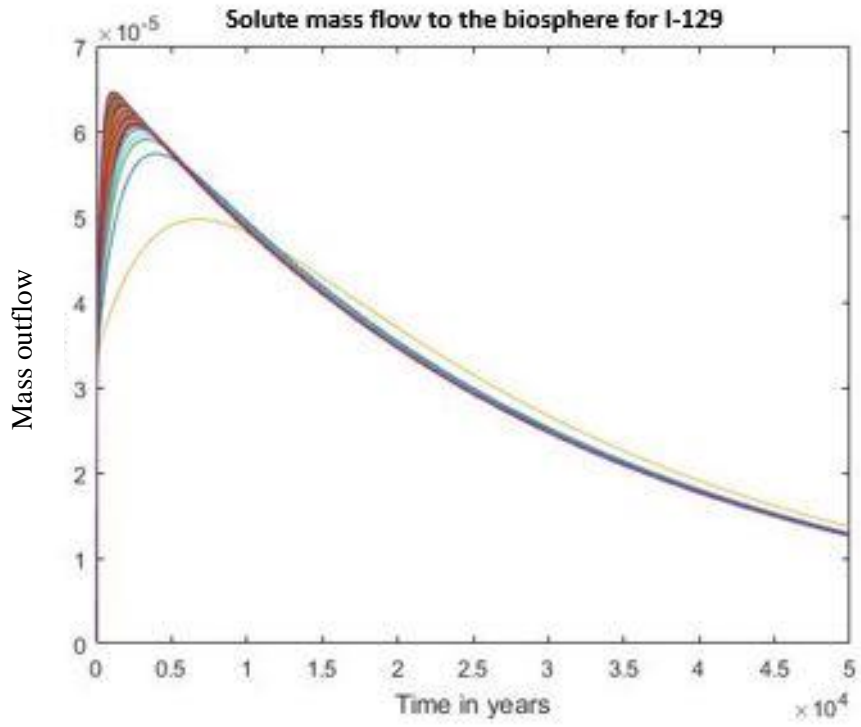


**Fig 4.11 Mass outflow of C-14 with random parameters for 200 runs**

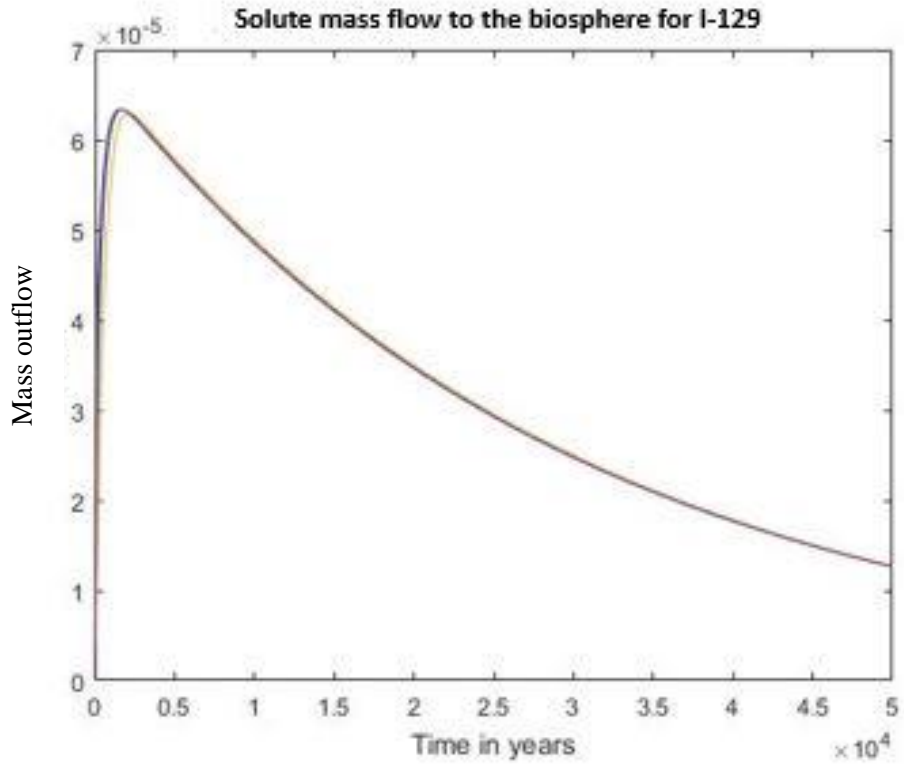
Fig 4.12, Fig 4.13, Fig 4.14 and Fig 4.15 gives the response curve for I-129 when the mass outflow terms from each of the barriers are incorporated with a 30% variance and the results are comparable with that of the C-14. Where the parameter  $\lambda_c$  (Fig 4.12) is the most sensitive of all the parameters and the response curve is unaffected by any changes in parameter  $\lambda_f$  (Fig 4.14) and  $\lambda_{tf}$  (Fig 4.15)



**Fig 4.12 Mass outflow with  $\lambda_c$  having variance of 30%**

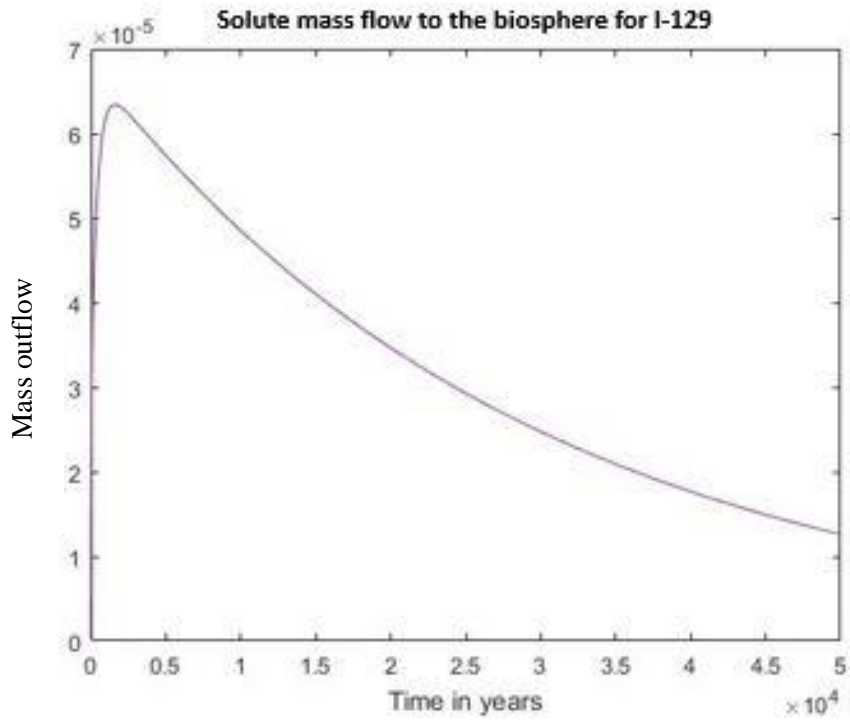


**Fig 4.13 Mass outflow with  $\lambda_{bt}$  having variance of 30%**



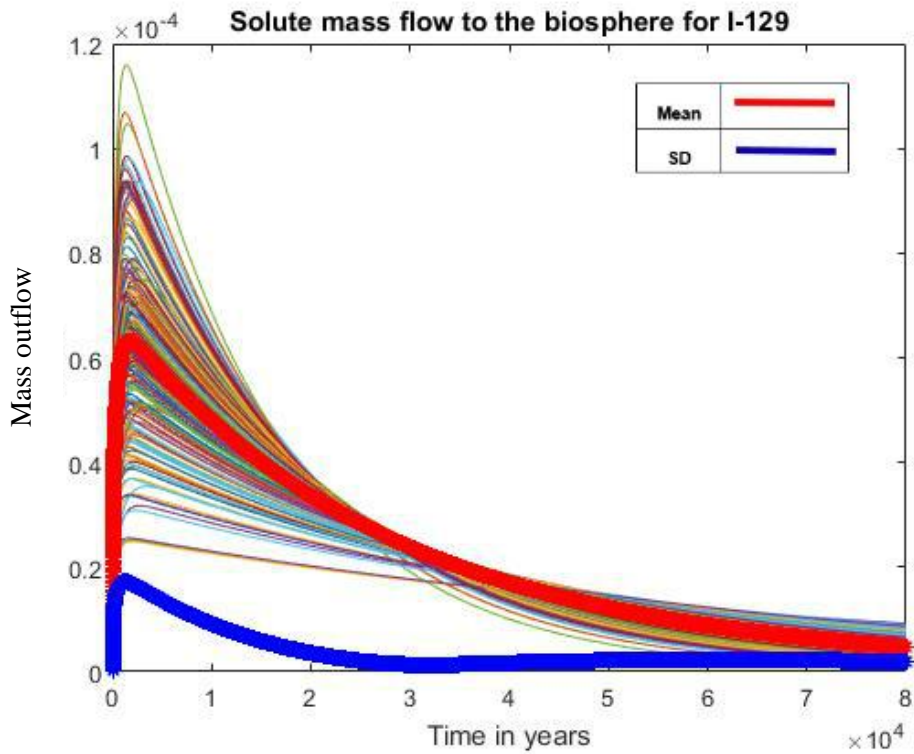
**Fig 4.14** Mass outflow with  $\lambda_t$  having variance of 30%





**Fig 4.15 Mass outflow with  $\lambda_{af}$  having variance of 30%**

Fig 4.16 gives the response curve for I-129 for 200 runs with 30% variance in all parameters. The model ran for a simulation period of hundred thousand years, after which there was no significant outflow. The peak value of mass outflow from the 200 runs is  $1.1594 \times 10^{-4}$  at 1379 years.



**Fig 4.16 Mass outflow of I-129 with random parameters for 200 runs**

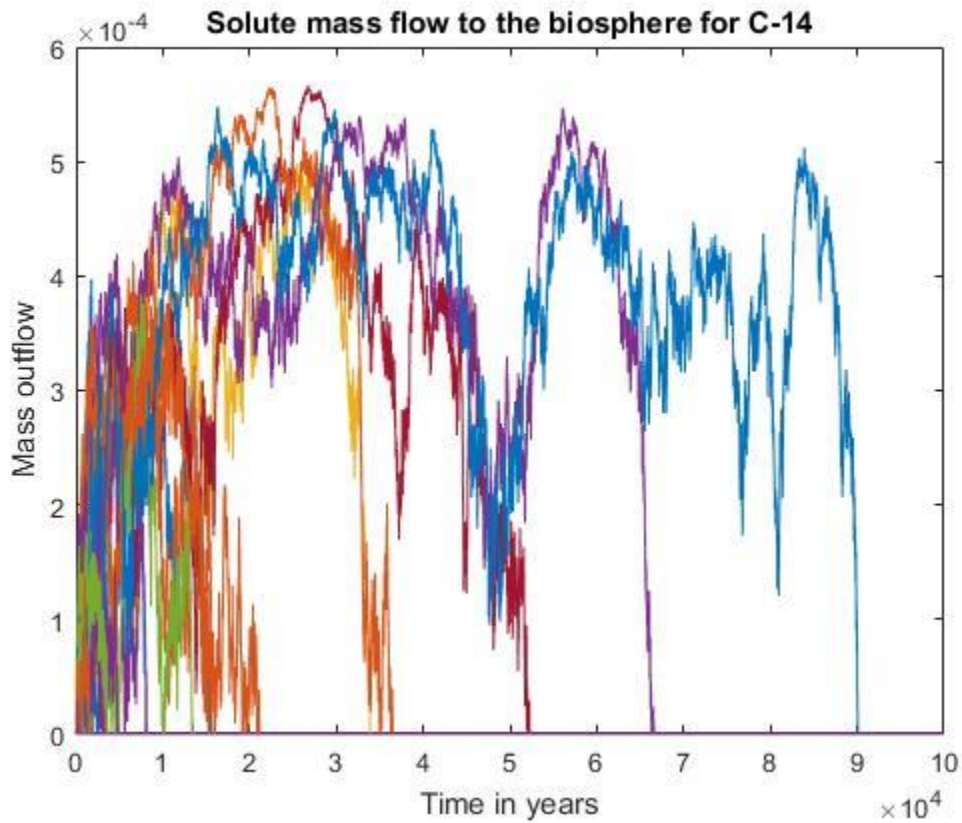
From Table 4.3 we can see that the mean of the peak value and the corresponding year is close to the results from the deterministic model results.

**Table 4.3 Numerical results from random parameter model**

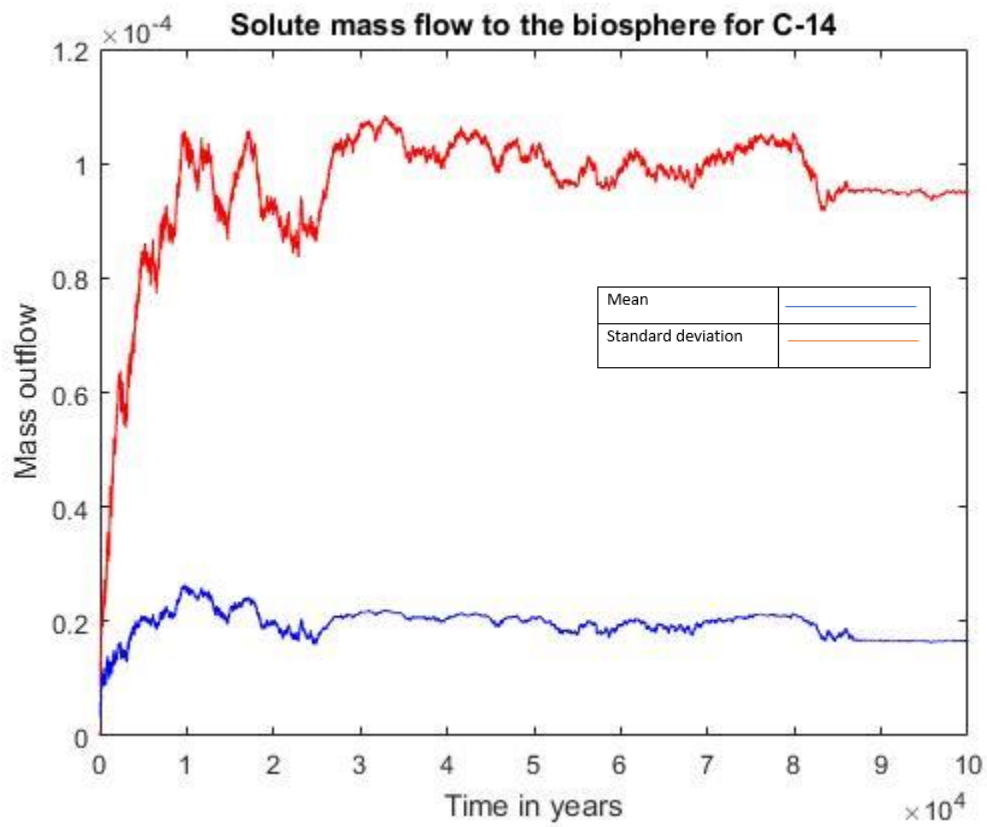
Isotope	Peak value of mass outflow (year <sup>-1</sup> )		Year	
	Mean	Standard Deviation	Mean	Standard Deviation
C-14	5.6069*10 <sup>-4</sup>	1.3828*10 <sup>-4</sup>	854.72	348.0968
I-129	6.3552*10 <sup>-5</sup>	1.7129*10 <sup>-5</sup>	1742	452.28

### 4.2.3 Stochastic model

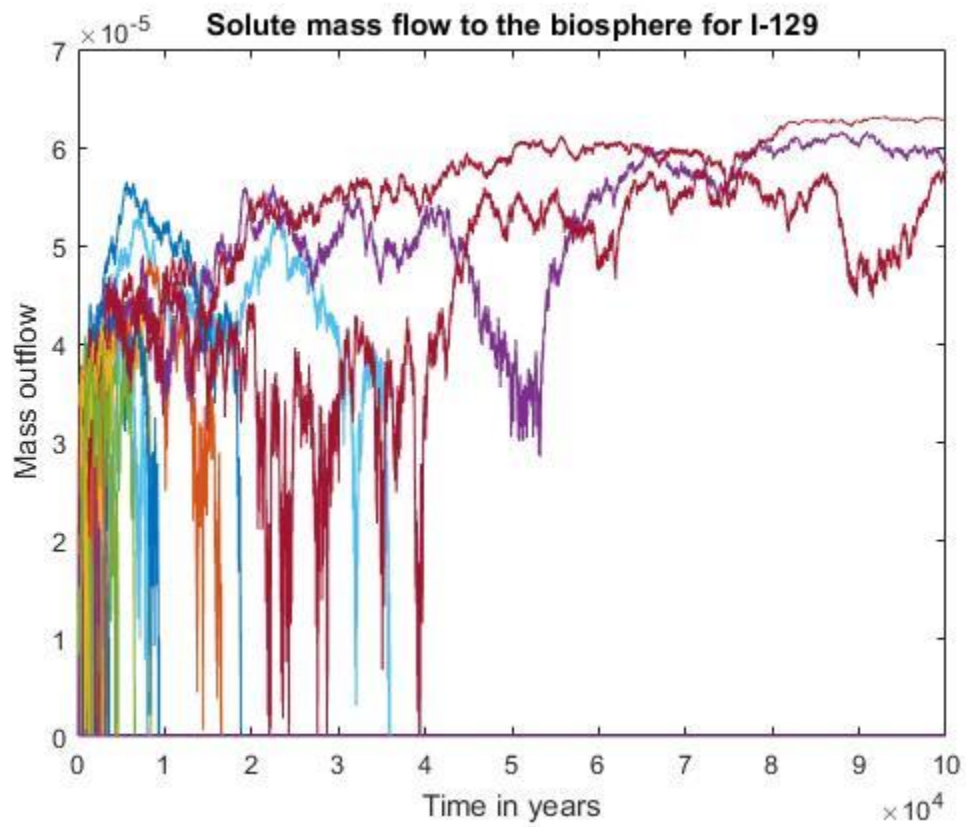
In this stochastic model, randomness of the parameter with respect to time is considered, and 200 samples of 10000 year simulation is run. The purpose of the stochastic model as to understand the post closure of the DGR since the time scale is very large and the event which could occur to cause a failure is uncertain. The rate outflow of mass of the entire system is represented stochastically for C-14 and I-129 and the results are shown in figures Fig 4.17 and Fig 4.19. The mean and standard deviation plots are in given in Fig 4.18 and Fig 4.20



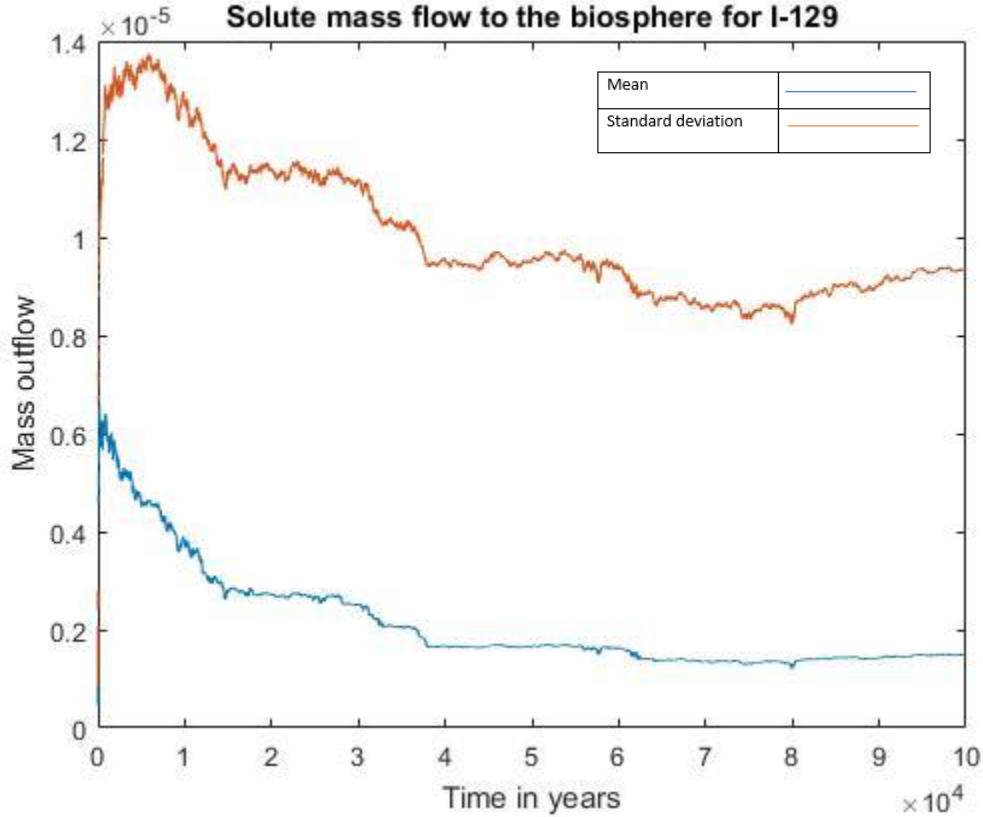
**Fig 4.17 Result of mass outflow from stochastic model for C-14**



**Fig 4.18 Mean and Variance curve for C-14 Stochastic model**



**Fig 4.19** Result of mass outflow from stochastic model for I-129



**Fig 4.20 Mean and Variance curve for I-129 Stochastic model**

#### 4.2.4 Radiation exposure calculation

The main aim of finding the decay or the activity of the radioactive species is to understand the impact it would have and ensure that it meets specifications set by international organizations nuclear safety organizations. The considered nuclide C-14 and I-129 have different decay mechanism, C-14 undergoes beta decay and I-129 undergoes gamma decay.

Table 4.4 gives the value of the dosage of radiation exposure from the decay of unit mass of C-14, since it is a beta decay it has very low energy any distance over 30-50cm the effect of the beta decay is not felt and the maximum distance is only in terms of few centimeters *Mook (1980)*.

**Table 4.4 Dose rate calculation for C-14**

Model	Maximum Mass outflow	Dose equivalent
	Year <sup>-1</sup>	rad/year at 20cm
Deterministic model	$5.7006 * 10^{-4}$	$1.98 * 10^{-18}$
Random parameter model	$5.6069 * 10^{-4}$	$1.95 * 10^{-18}$
Stochastic model	$2.63 * 10^{-05}$	$9.15 * 10^{-20}$

Table 4.5 gives the value of the dosage of radiation exposure from the decay of unit mass of I-129 which can be felt at 100 meters from the placement room.

**Table 4.5 Dose rate calculation for I-129**

Model	Maximum Mass outflow	Dose equivalent
	Year <sup>-1</sup>	mSv/year 100m
Deterministic model	$6.3378 * 10^{-5}$	$3.95 * 10^{-25}$
Random parameter model	$6.3552 * 10^{-5}$	$3.96 * 10^{-25}$
Stochastic model	$6.58 * 10^{-06}$	$4.11 * 10^{-26}$

The safety limit given by agencies like the International Atomic Energy Agency (IAEA), International Commission on Radiological Protection (ICRP) and European Union (EU) for exposure is 1mSv/year. The amount of fissionable material in a used fuel bundle is 0.74% and a used fuel bundle which weighs 19.2 kg will have 0.142 kg of fissionable material *Mroueh (2004)*. A mark II canister can hold 48 used fuel bundles. Hence, each canister has 6.816 kg of fissionable material. Assuming there are 100,000 canisters in a DGR and all of them fail the exposure will be in the order of  $2.6991 * 10^{-19}$  mSv/year which is still within the allowable limits.

## Chapter 5

### Conclusion

#### 5.1 Conclusion

With the increase in the use of nuclear reactors for power generation the need for isolating the highly radioactive waste has also increased. Countries which produce power using nuclear reactors would have to come up with a way to safely isolate the radioactive waste. The use of Deep Geological Repositories for storing highly radioactive waste is definitely a logical way for safely isolating them from the environment.

Based on the results from this thesis, we can conclude that water seepage into the repository is inevitable if the geological location has a water table nearby. The results of Case I show that the placement rooms will flood in about 5 years after saturation of the rock matrix and bentonite. Flooding of the placement room may lead to corrosion of the canister since water is the carrier of corrodents such as certain elements and bacteria, but Case II gives the results of exposure dosage for the worst-case scenario which assumes that the canister breaks and starts to leak radioactive substance as soon as the DGR is closed.

The results from Case II of the thesis shows that the multi barrier system such as the ones planned to be constructed in Sweden or Canada are safe even if there is a leakage from the canister due to any defects. From section 4.2.2 we know that the decay constant is the sensitive parameter in the model and the decay constant from canister to the buffer is the most sensitive parameter of them all. Decreasing the decay constant values can reduce the exposure dosage. However, the failure of all canisters is highly unlikely because the canisters undergo 100% testing as they are critical component in the system. The results in this thesis is given considering the worst-case scenario all canisters being broken, the radiation leak from the canister if any, will take several years to peak and the amount of radiation that may leak to the geosphere is well within the allowable radiation exposure. In reality, the exposure dosage will be lesser and will take longer since the model in Case II assumes that the canister fails as soon as the DGR is closed.



## **5.2 Future work**

For a more holistic approach to analyze the DGR system, a failure mode effect analysis can be done to identify the most critical components of the system. Since the canister is one of the most integral part of the system, corrosion analysis of the canister in the presence of bacteria, elevated temperatures and manufacturing defects can be analyzed. Stress analysis on the entire systems can be done considering the rock formation, decay heat and glacial pressure.

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