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Risk Evaluation of a Mercury Containment System

Cristian A. Ortez Garay
Florida International University, corte019@fiu.edu

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FLORIDA INTERNATIONAL UNIVERSITY

Miami, Florida

RISK EVALUATION OF A MERCURY CONTAINMENT SYSTEM

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Cristian Alejandro Ortez Garay

2011

To: Dean Amir Mirmiran
College of Engineering and Computing

This thesis, written by Cristian Alejandro Ortez Garay, and entitled Risk Evaluation of a Mercury Containment System, having been approved in respect to style and intellectual content, is referred to you for judgment.

We have read this thesis and recommend that it be approved.

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Georgio Tachiev

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Walter Tang

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Hector Fuentes, Major Professor

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The thesis of Cristian Alejandro Ortez Garay is approved.

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Dean Amir Mirmiran
College of Engineering and Computing

____
Dean Lakshmi N. Reddi
University Graduate School

Florida International University, 2011

ABSTRACT OF THE THESIS

RISK EVALUATION OF A MERCURY CONTAINMENT SYSTEM

by

Cristian Alejandro Ortez Garay

Florida International University, 2011

Miami, Florida

Professor Hector Fuentes, Major Professor

A probabilistic risk assessment model using GOLDSIM software was developed to evaluate the uncertainty of selected hydrological and soil parameters on mercury releases from a mercury containment system, which will be constructed within the Environmental Management Waste Management Facility in the Bear Creek Valley at the Oak Ridge Reservation in Tennessee. The main objective was to determine the concentrations and risk of exceeding the drinking water standard of mercury in a selected receptor well. A series of simulations were then conducted for various design periods, with emphasis on 10,000 years to determine those concentrations and risks. Experimental data for selected parameters such as dry bulk density, partition coefficient, and porosity and infiltration rate were represented by Probability Density Functions in support of Monte Carlo analyses. A sensitivity analysis showed that concentrations and risk are, for instance, most sensitive to porosity in the unsaturated zone. The simulations suggest that all herein estimates of concentrations and risks of mercury in drinking water should be well below established limits.

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AD	Advection Dispersion
BCV	Bear Creek Valley
CERCLA	Comprehensive Environment Response, Compensation, and Liability Act
CT	Contaminant Transport
CV	Coefficient Variation
EBCV	East Bear Creek Valley
EM	Environmental Management
EMWMF	Environmental Management Waste Management Facility
EPA	Environmental Protection Agency
HG	Mercury
LLRW	Low-Level Radioactive Wastes
MCA	Monte Carlo Analysis
MCL	Maximum Contaminant Level
MCLG	Maximum Contaminant Level Goal
NPDWRS	National Primary Drinking Water Regulations
NNSA/NV	National Nuclear Security Administration of the Nevada Office
NOAA	National Oceanic and Atmospheric Administration
NSC	National Security Complex
NT	Northern Tributary
NTS	Nevada Test Site
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reservation

PA	Probabilistic Assessment
PA/CA	Performance Assessment and Composite Analysis
PCB	Polychlorinated Biphenyls
PDF	Probabilistic Density Functions
PRA	Probabilistic Risk Assessment
RFD	Reference Dose
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
SDMP	Site Decommission Management Plan
SZ	Saturated Zone
TDEC	Tennessee Department of Environment and Conservation
USDOE	United States Department of Energy
USEPA	United States Environmental Protection Agency
USGS	United Stated Geological Survey
UZ	Unsaturated Zone
WAC	Waste Acceptance Criteria

INTRODUCTION

The Oak Ridge Reservation (ORR) was built in east Tennessee in 1942 as a part of the Manhattan Project during World War II. Four separate industrial plants were constructed in the race to develop the first nuclear weapon. The X-10 Plant (now known as Oak Ridge National Laboratory (ORNL)) was built as a pilot plant for the larger plutonium production facilities built in Hanford, Washington. The K-25, S-50 and Y-12 plants were constructed to separate uranium 235 (^{235}U) from the heavier ^{238}U using gaseous diffusion, liquid thermal diffusion process, and electromagnetic separation processes, respectively (Brooks & Southworth, 2011).

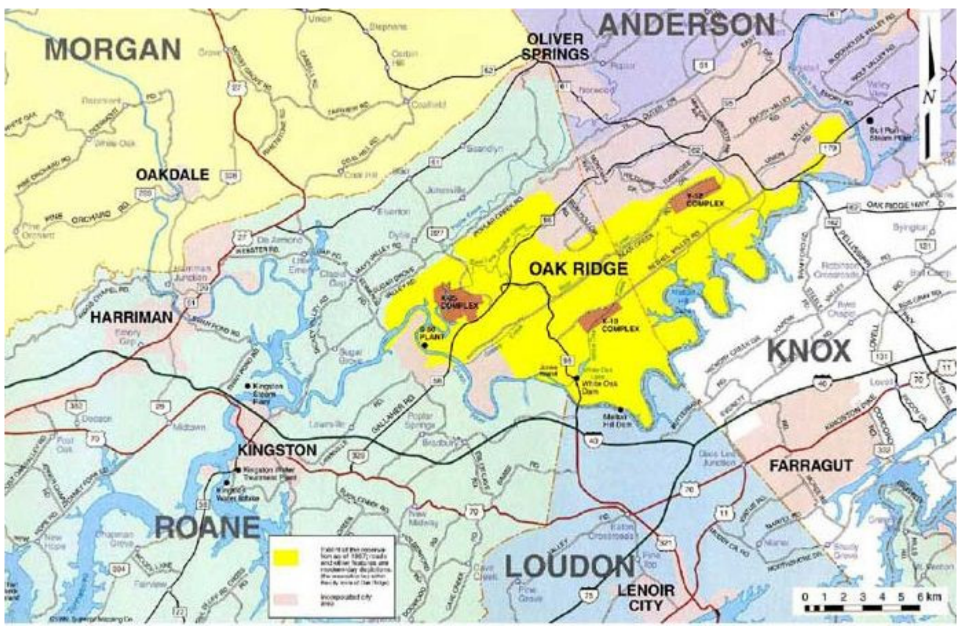

Figure 1 Location of the Oak Ridge Reservation (ATSDR, 2006)

Figure 1 shows a map of the location of Oak Ridge Reservation with the different complex facilities. Between the years of 1950 and 1963, about 11 million kilograms of mercury (Hg) were used at the Oak Ridge Y-12 National Security Complex (Y-12 NSC) for lithium isotope separation processes (Brooks & Southworth, 2011). According with

Brooks & Southworth about 3 percent of the mercury was lost to the environment including air, soil, and rock under the facilities.

Mercury is a pollutant of global concern, which is largely due to its potential for biological transformation into harmful forms, bioaccumulation, and biomagnifications through the ecological food chains (USEPA, 1997). Mercury contamination is present in the Y-12 NSC watershed and has been identified, as a key contaminant in soil, sediment surface water, groundwater, buildings, drains, and sumps. Most of the contamination around Y-12 NSC is restricted to the upper 10 feet of soil and fill (Han et al., 2006). To remedy and contain the contamination of mercury in the surroundings areas of Y-12 NSC, a new mercury containment system has been proposed. The designated area to host this new containment system is the Environmental Management Waste Management Facility (EMWMF) (USDOE, 1998).

The EMWMF is a containment system facility, which is authorized by the United States Environmental Protection Agency (USEPA) and the Tennessee Department of Environment and Conservation (TDEC) for long-term storage of wastes generated by environmental restoration activities. The environmental restoration activities are being conducted at the United States Department of Energy's (USDOE) Oak Ridge Reservation as part of the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) cleanup of the ORR (Benson, 2008). EMWMF is approved to receive low-level radioactive wastes (LLRW), hazardous wastes, and mixed wastes (Benson, 2008). All the operation activities performed at the EMWMF are designed to prevent the release of contaminants into the environment and to meet regulatory guidelines. Operating controls minimize the release of contaminants into the air through dust control management, into surface water through storm controls, and into groundwater through the design of and operation of a liner and leachate collection system. Figure 2 shows a plan view of the EMWMF with the existing cells and the perimeter drainage channel (USDOE, 2008).

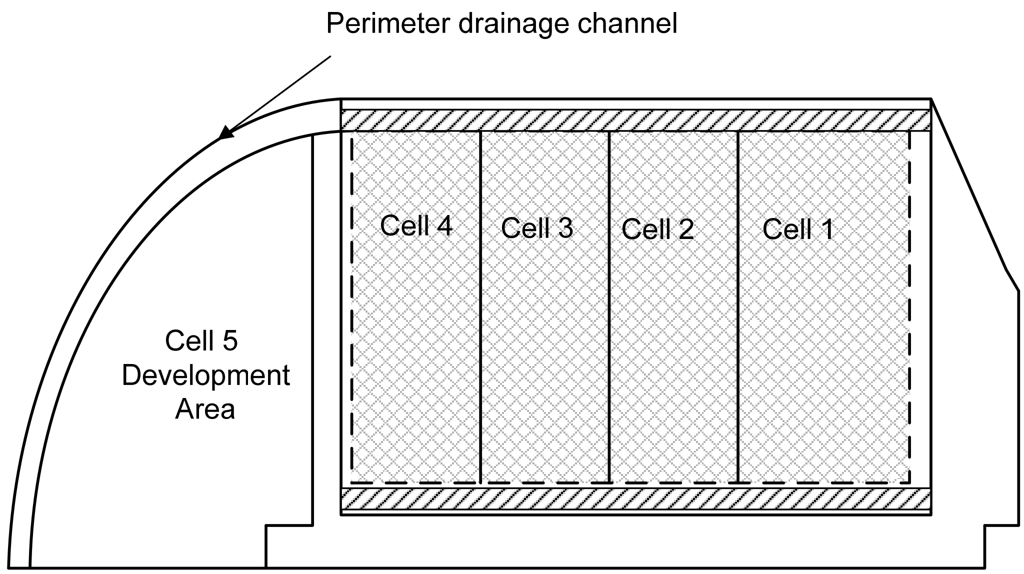

Figure 2 Plan View of EMWMF showing existing cells

The EMWMF site is located in a ridge within the East Bear Creek Valley (EBCV) and west of the Y-12 Main Plant area (Corpstein, 2003). The EBCV site is relatively flat at the south with a series of knolls to the north, and is transected by Bear Creek North Tributary (NT-4) (USEPA, 1999). At the nearly flat valley floor, the groundwater table is near the ground surface. On the valley slopes, moving upgradient to the ridge crest, the groundwater table can be deeper than 15 meters. Groundwater movement is relatively slow with discharge to Bear Creek and its tributaries.

At the location of the EMWMF, contaminants may leak from the containment system to the unsaturated zone, then mix with groundwater and travel downstream to extraction well GW-904, which could potentially be used by humans. By specifying actual or hypothetical well locations, the peak concentrations of contaminants in the groundwater can be determined for a given configuration of the disposal system. Target receptors, such as humans that consume water drawn from a well, can be used to estimate the potential doses and risk of the presence of contaminants in the groundwater (USDOE, 1998).

Throughout the assessment of on-site waste management options, many assumptions were made to accommodate uncertainties in waste inventory, physical and environmental data, pathway analysis, and land use considerations. The hypothetical receptor scenario used for the risk assessment of the disposal facility needs to satisfy the risk/toxicity criteria for all radiological and chemical constituents with a risk $\leq 1x10^{-5}$ for a post closure period $\leq$ 1,000 years and a risk of $\leq 1x10^{-4}$ for a post closure period > 1,000 years. The receptor location is a major assumption for this risk assessment, as currently residential use of groundwater or surface water in Bear Creek Valley is not allowed. Future land use plans have been drafted, which specify releasing the western portion of the valley for residential use (DOE 1998c). Because the disposal facility would be located among the other CERCLA remediated sources, it would be constructed in a future DOE-controlled Brownfield area and located at least 1.8 km (1.1 miles) upstream of the nearest public receptor permitted by those plans. Well GW-904 is located one meter southwest of the mercury containment system. This well was conservatively chosen based on its proximity to the facility, and analysis conducted by the DOE and TDEC on site topography, geology and preliminary groundwater impact modeling (USDOE, 1998). In addition this well is within the area of influence of any groundwater impacts caused by the operation of the disposal facility.

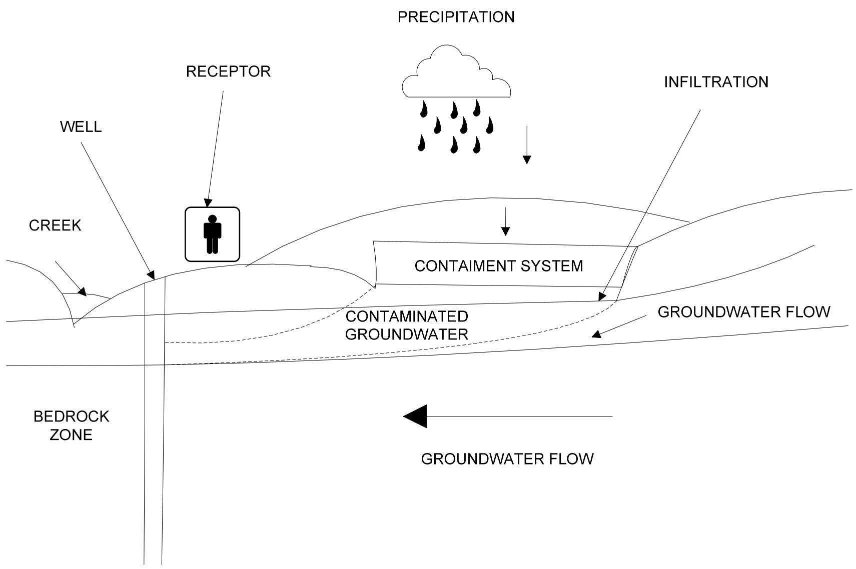

Figure 3 Schematic of a mercury containment system
and hypothetical leakage pathways

Figure 3 shows the schematic of a typical mercury containment system and hypothetical impact on groundwater when leakage occurs. The principal processes that influence transport behavior of mercury in groundwater are advection, dispersion, sorption, and chemical transformation (Devinny et al., 1990).

DOE's Order 435.1 (Hiergessel & Taylor, 2008) provides performance objectives for disposal of low-level radioactive waste (LLW) at DOE sites, which include a probabilistic assessment (PA) required to evaluate all low-level radioactive waste disposal facilities at DOE sites. According to DOE's Order 435.1, the performance assessment is required for all new mercury containment systems. The purpose of the performance assessment is to determine the potential risk of impact on the public and the environment (Ho et. al., 2002). Furthermore, DOE's order M 435.1-1, defines performance assessment studies as analysis of radioactive or chemical waste disposal facilities to demonstrate reasonable expectations that the performance objectives established for the long-term protection of the public and the environment will not be exceeded after closure of the facility. One of the PA requirements of DOE Order 435.1 is to evaluate the sensitivity and uncertainty in achieving the performance goals and measures. A probabilistic risk analysis methodology was developed to facilitate the quantification of risks associated with complex engineered systems and uncertainty in selected parameters. In general, the probabilistic methodology is particularly well suited to analyzing the frequencies of extremely rare events; however, a probabilistic risk analysis model is considerably more complex than traditional single-point estimates using deterministic models (Molak, 1996).

Recent analysis of the performance of disposal cells at the EMWMF for radioactive and hazard constituents have primarily relied on deterministic models of flow and transport processes and have ignored the uncertainty of important environmental parameters. These parameters include variables related to the hydrological cycle and soil's physical properties (Johnson & Urie, 1985) that directly affect the long-term performance of the containment system (Ho et al., 2002). In general, the time period used for probabilistic models to evaluate peak concentrations of radioactive and hazard constituests is 10,000 year (USDOE, 2010).

The performance of cells 1-6 of the containment facility and the potential exceedance of the waste acceptable criteria (WAC) were analyzed for 13 radioactive and 123 hazardous constituents. WAC specify concentration limits of radionuclides and hazardous chemicals for various waste forms such as soil, solidified and stabilized wastes and debris such that long term human and environmental risks do not exceed the risk and toxicity goals of each constituent. These limits depend in part on the receptor, location, exposure scenario, and disposal cell design (USDOE, 2010).

A risk performance assessment for the radioactive and hazardous constituents was conducted using combinations of risk analysis models such as PATHRAE-RAD and PATHRAE-HAZ (RAEC, 1995) to calculate the concentration in the groundwater, the risk and dose of the different constituents. A monitoring well near the facility, GW-904, was designated to be used by a hypothetical receptor, a resident farmer who used the water from the well for human consumption. The PATHRAE-HAZ model, which is a deterministic performance assessment program for the land disposal of hazardous chemical wastes, was used for hazard constituents like benzene, dieldrin, tin, among others, for the first 100,000 years after closure. This study took into account many hazardous chemicals for the analysis, however mercury was ommited. The model simulations indicated that the resultant risk and doses to the receptor would not exceed the current WAC criteria for any of the constituents.

Beyond the deterministic models, the risk related to mercury containment at EMWMF can be determined using risk probabilistic software. The Contaminant Transport (CT) module in GOLDSIM software is an extension of the GOLDSIM general program, which provides probabilistic simulations of the release and transport of a mass of contaminants within a complex engineering environmental system, such as a containment system, to the unsaturated and saturated zone. The fundamental output produced by the CT module consists of predicted concentrations within environmental medium, such as soil, groundwater, and air, throughout the system (GOLDSIM, 2010). The concentrations in environmental medium can be converted to receptor doses and health risks by assigning appropriate conversion factors and equations (Kossik & Miller, 2004). The module offers the ability to input key hydrological and soil parameter values to create a probabilistic risk assessment model. This model is capable of simulating the transport of contaminants in the subsurface using probability distributions for the uncertainty of key parameters in an advection-dispersion module (GOLDSIM, 2010).

The USDOE National Nuclear Security Administration of the Nevada Office (NNSA/NV) operates and maintains two facilities on Nevada Test Site (NTS) that dispose defense generated low-level radioactive waste (LLW), mixed radioactive waste and classified waste. The Nevada PA maintenance program has the primary goal to ensure that the conclusions of the performance assessment and composite analysis remain valid over the operational life of the LLW disposal facility as well the post closure period (Crowe et tal., 2002). A range of well-documented commercial computer software programs were examined for application to probabilistic performance assessment (PA) modeling. Based on an examination the GOLDSIM Contaminant Transport Module extension was selected to be used in the PA maintenance program. The primary strengths of GOLDSIM include the following:

I. It has been designed as a fully probabilistic computer program.

II. It provides integration PA applications, and the software contains modules designed for probabilistic modeling of the multiple components of a waste disposal system.

III. The GOLDSIM software has been used for multiple national and international performance assessment studies, including the total system performance assessment studies of underground disposal of high level radioactive waste by the Yucca Mountain project (USDOE, 2001).

IV. The software code is well documented (McGrath & Beckham, 2001).

The NNSA/NV program is in the process of converting the deterministic PA/CA for the facilities into integrated probabilistic models (Crowe et tal., 2002).

One of the multiple applications of the GOLDSIM contaminant transport module is the capacity to operate as an integrator that samples the uncertain distributions of selected input parameters such as bulk density of the soil, porosity of material, and distribution coefficient. These parameters are linked with the Breach, Leach, and Transport-Multiple Species (BLT-MS) program (NRC, 1989) to do a probabilistic risk assessment of the subsurface low-lewel waste disposal facility. The results show that GOLDSIM can be successfully integrated into another program using its linkage capabilities (Mattie et. al., 2007).

The GOLDSIM contaminant transport module for the mercury containment system can simulate one-dimensional advection-dispersion transport of contaminants in the groundwater. In order to built a model, which represents a specific situation, such as the release of mercury form a containment system to the groundwater, GOLDSIM has to connect different elements. The key elements for the contaminant transport module in GOLDSIM are listed in Table 1. The elements were linked multiple times in the model to create a complex numerical simulation of the groundwater contaminant transport.

Table 1 List of key elements included in the transport module

GOLDSIM elements	Task
Cell	Containment System 1-D Advection
Pipe (UZ)	Unsaturated zone 1-D Advection-Dispersion
Pipe (SZ)	Saturated zone 1-D Advection-Dispersion

1.1 Research objectives and justification

The main purpose of this study is to perform a probabilistic risk assessment analysis and to evaluate how the stochastic distribution of selected soil parameters, such as dry bulk density, porosity, partition coefficient and infiltration rates, have an impact on the release and transport of mercury-contaminated water at a containment system facility. This methodology provides a better understanding of the impact of modeling parameters on mercury concentration in the groundwater compared to a deterministic model, such as the hydrological evaluation of landfill performance (HELP) (Schroeder et al., 1994).

A contaminant transport model using GOLDSIM was built in order to predict the transport of mercury from the containment system to the subsurface zone. In addition, a risk probabilistic assessment evaluation of the model was performed to include the extraction of drinking water from the well, calculations of the dose, and risk for a period of 10,000 years. Monte Carlo simulation was applied with the purpose of understanding how uncertainty in these selected parameters has an impact on the peak concentration of contaminated groundwater, and therefore an impact on the risk to and dose for the human receptors. Table 2 lists the steps of the development of this model.

Table 2 Steps for the development of the GOLDSIM probabilistic model

Step	Description
1	Develop a contaminant transport module in GOLDSIM
2	Characterize input parameters
3	Run deterministic calculations
4	Develop a risk assessment model
5	Develop probabilistic distribution for selected uncertain parameters
6	Perform simulations and analysis of selected parameters
7	Perform calculations and sensitivity analysis of the probabilistic simulations
8	Interpret and document results

This plan provides the critical data for the transport of mercury from the containment system using a one-dimensional advection-dispersion model. The purpose of the probabilistic analysis is to provide added insight into the release pathway mechanisms, help identify the most important parameters, and either question or lend confidence to the deterministic results as compared with the facility disposal limits (Hiergessel & Taylor, 2008).

1.2 Site description

The climate of the Oak Ridge region is broadly classified as humid and subtropical. The region receives a surplus of precipitation compared to the level of evapotranspiration that is normally experienced throughout the year. Evapotranspiration in the Oak Ridge area has been estimated at 74-76 cm or 55-56% of annual precipitation (TVA 1972, Moore 1988, & Hatcher et al., 1989). The 30-year annual average precipitation (1976–2005) is 1374.3 mm, including about 27.4 cm of snowfall (NOAA 2006). The bedrock on the Oak Ridge Reservation (ORR) ranges in age from 250 to 550 million years. In general, the valleys in this area are underlain by bedrock formations predominated by siltstones and limestones, including the Conasauga Group and the

Chickamauga Group. The Chickamauga group underlies Bethel Valley and contains fractured bedrock, predominantly siltstone, shale, sandstone, and thinly-bedded limestone. The most significant water flow occurs within a depth of 1-3 m, referred to as the storm flow zone, which approximately corresponds to the root zone of the vegetation. However, groundwater flow, and contaminant transport occurs at a depth ranging between 10-50 m (Hatcher et al., 1992). The hydrologic units in the ORR include the Knox aquifer, which includes the Maynardville Limestone and is highly permeable, and the aquitards, which consist of less permeable geologic units.

Knowledge of the ORR geology is necessary to provide detailed information of factors controlling groundwater flow and the data required to develop a contaminant transport model. The proposed disposal facility is located in the upper section of Bear Creek Valley.

Figure 4 shows the location EMWMF on the Oak Ridge Reservation. The elevation of the valley floor ranges from about 287 to 305 m (940 to 10000 ft).

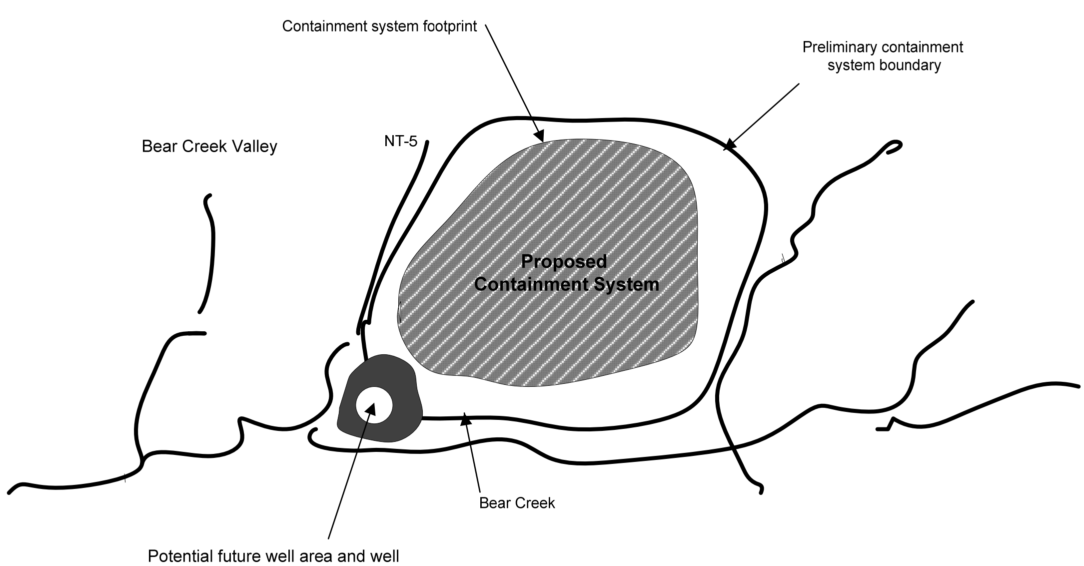

Figure 4 Location of the EMWMF on the Oak Ridge Reservation

Small-scale geologic structures, such as fractures and solution features, are a major factor in groundwater movement through the formation underlying the proposed disposal facility. These bedrock characteristics provide the pathways for groundwater flow through geologic formations that have little primary porosity and permeability. Fractures are well developed in bodies of rocks that are formed with definable units based on their own geological properties and are the most common structures (Hatcher et al., 1992). The orientations of well-connected fractures are mainly parallel to geologic strike and increase the effect of anisotropy, which is caused by layering, resulting in dominance of strike parallel groundwater flow paths. Fracture aperture width and frequency generally decreases with depth in all formations and thus restricts the depth of active groundwater circulation. The Maynardville Limestone and overlying Knox Group exhibit widespread evidence of dissolution, which is manifested as enlarged fractures and well-developed, well-connected cavity systems.

The unconsolidated materials underlying bedrock at the proposed disposal facility location include mostly saprolite, which is a mixture of residuum and bedrock remnants, weathered bedrock, and fill associated with previous disposal activities.

Within Bear Creek Valley, the majority of groundwater flow is hypothesized to occur within the upper 30 m (100 ft) of the aquifer system. The occurrence and movement of groundwater in the bedrock aquifer system is closely related to the presence of bedding planes, joints, fractures, and solution cavities. In general, groundwater in bedrock occurs under water-table conditions but it becomes increasingly confined with depth. Downward recharge to the groundwater system occurs along the flanks of Pine Ridge and Chestnut Ridge. Because of the orientation of fractures, hydraulic conductivity is anisotropic and is highest along geologic strike. This anisotropy causes groundwater to flow primarily along strike (i.e. east to west). Because of this along-strike flow, a large portion of the shallow groundwater discharges into the tributaries (e.g. NT-5, for groundwater flowing beneath the proposed disposal facility location) and eventually flows into Bear Creek.

Groundwater movement within the siliclastic units is dominated by fractured flow (Solomon et al., 1992). More than 95 percent of the flow occurs through the shallow interval. Although only limited hydraulic testing has been done at the proposed disposal facility location, many hydraulic tests (e.g. pumping, slug, packer, bailer, and tracer tests) have been conducted in geologic units within Bear Creek Valley. The data was compiled and summarized by the Jacobs Environmental Management (EM) Team during Bear Creek Valley regional groundwater flow model development (DOE 1997). Hydraulic conductivities calculated from the tests range over five orders of magnitude, from $1x10^{-3}$ to $1x10^{-8}$ cm ($1x10^{-5}$ to $1x10^{-10}$ ft)/second within each hydrostratigraphic unit. In general, the wide range in hydraulic conductivity values is due to the heterogeneous distribution of fractures and the scale at which many of tests were performed. The relationship between hydraulic conductivity and depth shows a weak correlation between hydraulic conductivity and depth, where the average hydraulic conductivity in the first 30 m (100 ft) appears to be higher than the hydraulic conductivity in the deeper portions of the bedrock aquifer system. This is expected in bedrock aquifer system where the size and abundance of fractures usually decreases with depth.

Various aquifer tests indicate that the Bear Creek's Valley hydrogeologic units are not isotropic. They behave instead as anisotropic systems in all three dimensions, evidenced by the elongated drawdown along strike direction observed during pumping tests and the spatial distribution of contaminant plumes. The anisotropic nature of hydraulic conductivity associated with the bedrock underlying Bear Creek Valley is apparently caused by the orientation and intersection of fracture, join, and bedding planes. Vertical hydraulic gradients appear to be predominantly upward in the siliclastic units of the Conasauga Group. The prominence of a vertically upward gradient is attributed to the anisotropy of the formations and connections with the recharge area located along Pine Ridge. In the Maynardville Limestone, the distribution of vertical gradient varies (USDOE, 2010).

RISK ASSESSMENT GENERAL CONCEPTS

Previous risk and dose assessment analysis for a containment system at the EMWMF facility was performed for the first 10,000 years after closure and took into consideration a range of radiological and hazards constituents, which were hypothesized as derived from the consumption of drinking water from a well located near NT-5 between the EMWMF and Bear Creek (USDOE, 2010).

The peak risks and doses were calculated using the PATHRAE program (Rogers and Associates Engineering (RAE) 1995a and 1995b). PATHRAE is used to calculate the annual dose for the pathway of groundwater migration with discharge to a well. This pathway consists of downward migration of waste components by advection or as a result of dissolution in percolating precipitation. The waste components move downward through the unsaturated zone to an aquifer beneath the disposal site. In the aquifer, the waste components are transported by advection and dispersion and the contaminated aquifer water is withdrawn from a well (EPA, 1987). The DOE has reported that for the majority of radiological and hazards, the analyzed contaminants will not exceed the current WAC criteria. In addition, they reported that most of the risk and doses to the receptor comes from contaminated drinking well water. Therefore, any major reduction in constituent's concentration in the groundwater at the well will greatly reduce the projected risk and doses (USDOE, 2010). Nevertheless, in the 13 radiological and the 123 hazards constituents present in the report, a risk assessment analysis for mercury was not performed. Therefore, a new risk analysis was required for the mercury containment system.

Moore et al, 1998, developed a probabilistic risk assessment of the effects of methyl mercury and PCBs on Mink and Kingfishers along the East Fork Poplar Creek in Oak Ridge, Tennessee, with the purpose of estimating the risks posed by methyl mercury and PCBs to two piscivorous species: mink and belted kingfishers. The authors used Monte Carlo simulations to estimate the intakes of each contaminant by each species and subsequently integrated the resulting distributions with their respective dose-response curves to estimate the associated risks. The Monte Carlo analyses for exposure combined the input distributions. Each analysis included 10,000 trials and Latin Hypercube Sampling to ensure adequate sampling from all portions of the input distributions. The results indicated that methyl mercury poses a moderate risk to female mink (24% probability of at least 15% mortality) and kingfishers (50% probability of at least a 12-18% decline in fecundity depending on location). Furthermore, the study concluded that given the serious risks posed by methyl mercury to mink and kingfishers, the next step is to evaluate possible remedial options that could be used to reduce risks to acceptable levels.

Saponaro et al., 2005, developed a risk assessment procedure to identify the remediation actions that may be adopted at a mercury-contaminated site. Analytical and numerical fate and transport modeling tools were used to locate digging zones in contaminated subsoil, to reduce the possible groundwater contaminant loading, and to avoid exceeding concentration limits. In general, site characterization is a critical factor in defining the conceptual model and in assessing risk; it is designed to acquire both data about the soil and groundwater contaminants, and parameter values for fate and transport modeling of contaminants through the environmental matrices (Ferguson et al., 1998).

The Saponaro et al., 2005 study concluded that even the most abundant mercury species in soil are poorly leachable under the site-specific environmental conditions. In general, human health and environmental risk assessments for metals are difficult to estimate because environmental behavior and toxicity depend on metal chemical forms and soil properties, such as pH value and redox potential, and these properties greatly vary in the environment. Specific tests for studying metal mobility and availability are required to provide data about the total concentration in soil (Evan, 1989; Holm et al., 1998; Ma & Rao, 1997). In risk assessment procedures, metal mobility in soil is taken into account by its distribution coefficient Kd; this factor relates the chemical sorbed to the soil solid phase per unit of mass to the concentration of chemical remaining in the soil solution at equilibrium.

The U.S. EPA reports a solids-water distribution coefficient (Kd) for the elemental mercury of 1000 ml g^{-1} (USEPA, 1997). The mercury contamination remediation was performed using the RISC 4.0 (Waterloo Hydrogeologic, 2001) program to determine the fate and transport of mercury. The soil dry bulk density, distribution coefficient, and soil effective porosity were parameters affecting the mobility of mercury concentration in the unsaturated zone.

A study in Nevada of mercury contained in buried landfill waste reveals a potential lateral migration of elemental Hg through the unsaturated zone (Walwoord et. al., 2008). The study concluded that transport of elemental Hg through arid unsaturated zone is a viable long-distance pathway for mercury migration from landfills. Future work is needed to better understand controlling processes and to quantify parameters.

The probabilistic modeling approach has been widely used to perform risk assessments for contaminated sites (USEPA, 1997; Hope & Stock, 1998; Slob & Pieters 1998; Chang, 1999; USEPA, 2001; Liu et al., 2004; Li et al., 2007). Nevertheless, only a few models, including GOLDSIM, use Monte Carlo simulations and stochastic analysis applied to contaminant fate and transport.

The stochastic approach in modeling groundwater flow and solute transport is related to the aquifer properties and the parameters that influence flow and transport as random. The randomness reflects the uncertainty of their values; the most common example is the hydraulic conductivity K among other properties of heterogeneous formation such as natural recharge aquifer geometry. The field data based on measurements are generally scarce and permit estimating values in statistical terms only. The probabilistic density function (PDF) of properties and parameters serves as an input to the quantitative modeling of flow and transport, resulting in stochastic differential equations for the dependent variables such as contaminat concentration. As a result, the contaminant concetration can also be described statistically by their PDF distributions or in a more restricted way by a few variables such as mean and the standard deviation (variance). Therefore, considering that forecast calculations are subjected to uncertainty, probabilistic risk assessment is the appropriate approach; which is in contrast with the traditional deterministic modeling of groundwater flow and transport (Dagan, 2002).

The Savannah River National Laboratory (SRNL) has developed a hybrid approach to performance assessment modeling using a multi-dimensional modeling platform (PORFLOW) to develop deterministic flow fields and perform contaminant transport. The GOLDSIM modeling platform is used to develop the sensitivity and uncertainty analyses. (Taylor & Hiergesell, 2011).

The approach at the Savannah River Site (SRS) was to use PA's to establish facility disposal limits based on the maximum permissible exposures to hypothetical individuals over 1000 year PA period of compliance. Limits are based on the highest exposure received by an individual through any of the analyzed pathways. According to several studies, the analyses are typically carried out for 10,000 years and longer in order to determine when a peak dose would occur (Hiergessel & Taylor, 2008).

An assessment of mercury release in the Savannah River Site Environment from the solid waste disposal facility (SWDF) was performed. The SWDF have approximately 10,000 kg of mercury. Orebaugh and Hale (1976) made a mathematical model of the risk from mercury in the SWDF and seepage using 9,000 Kg of mercury as the source term. Orebaugh and Hale concluded that a mercury flux of approximately 219 mg/hr might enter the water table and travel horizontally from the SWDF. As a worst case, this flux could contribute approximately 0.0002 ppm to the stream (Orebaugh & Hale, 1976). This concentration is below the drinking water standard of 0.002 ppm (USDOE, 1994).

GOLDSIM was used to develop a screening PA model of a reference low-level radioactive waste (LLRW) disposal facility to evaluate the risk and uncertainties associated with the containment system of depleted uranium as low level waste (Pinkston et. al., 2009). GOLDSIM have proved to be reliable software to performed probabilistic calculations to evaluate the radiological risk to future residents near or on the land overlying the containment system waste (Pinkston et. al., 2009).

TRANPORT AND FATE OF MERCURY IN SUBSURFACE

Due to its chemical properties, environmental mercury is thought to move through various environmental compartments, possibly changing form and species during this process (USEPA, 1997). Mercury occurs in several oxidation states, including Hg^0 (elemental), Hg_2^{2+} (mercurous ion), and Hg^{2+} (mercuric-Hg (II)). The properties and chemical behavior of mercury strongly depend on the oxidation state. Mercury solubility will be dictated by the speciation of the mercury in the system being observed.

Elemental mercury is an extremely dense liquid (13.595 g/cm^3) practically insoluble in water. Hg^0 it behaves as a dense no aqueous phase liquid, flowing downward under the influence of gravity through porous materials until it reaches an impermeable surface on which it will pool. It is volatile at normal earth surface temperatures and will vaporize in the unsaturated zone and dissolve in water, with the equilibrium solubility of 25 µg/L in a closed system. The amount present in water open to the atmosphere will likely be much lower because of its volatility. Mercury has a strong affinity for sulfide (as

Hgs) and selenide (as HgSe) ions. Most of the environmental concern is associated with methyl mercury because it bioaccumulates in the food chain and can cause neurological injury and death. Low pH and reducing conditions associated with a source of dissolved organic carbon provide conditions for the formation of methyl mercury.

Nearly all of the mercury found in all environmental medium, with the exception of the atmosphere biota, is in the form of inorganic mercuric salts and organomercurics. (USEPA, 1997). Due to the affinity of inorganic mercury for sulfur containing compounds in soils, it tends to form complexes primarily with soil organic matter and to a lesser extent to mineral colloids. These processes limit mercury mobility in the soil.

Mercury can enter the freshwater in different forms, organic or inorganic, wet or dry, and from different sources, a deposition from the atmosphere, or as part of the runoff "bound to suspended soils/humus or attached to dissolved organic carbon" (USEPA, 1997), or from groundwater because of leaching from soil.

The leaching of contamination depends on several factors. The principal processes that influence the transport behavior of contaminants in groundwater are advection, dispersion, sorption, and transformation (Bedient et. al., 1994). For dissolution of a heavy metal reacting with a solid phase, the process is know as adsorption-desorption in the sediment phase, adsorption-desorption in the water phase. Adsorption and desorption are processes by which the metal is transferred between solute and solid phases. Advection and dispersion describe the rate of movement and dilution of contaminant or solute. Advection is the transport of solute by the volume groundwater flow. Dispersion is the spreading of the plume that occurs along and across the main flow direction due to aquifer heterogeneities at both scale, pore scale (small) and regional scale (macroscale). Dispersion tends to increase the plume uniformity as it travels downstream. Factors that contribute to dispersion include faster flow at the center of the pores than at the edges. Sorption, or the partitioning of the contaminant between the liquid and solid phases, results in a decrease in concentrations in the water without changing the total mass of the compound, and in the retardation of its movement relative to groundwater flow (Delleur, 2000). Sorption refers to the exchange of molecules and ions between the solid phase and the liquid phase. It includes adsorption and desorption. Adsorption is the attachment of molecules and ions from the solute to the rock material. Adsorption produces a decrease of the concentration of the solute or, equivalently causes a retardation of the contaminant transport compared to the water movement. Desorption is the release of molecules and ions from the solid phase to the solute (Delleur, 2000).

The relationship between the contaminant concentration in the adsorbed phase and in the water phase is called a sorption isotherm. The adsorption causes retardation in the migration of contaminants compared to the advection. The contaminant transport gets more retarded as the fraction adsorbed increases. The partition coefficient defines the retardation factor, Equation 1.

$$Rf = 1 + \frac{k_d \rho_b}{\theta} \qquad (1)$$

Where Rf is the retardation factor, Kd is the partition coefficient, ρb is the bulk density, θ is the porosity. If the Rf is a larger value, the contaminants will delay behind the movement of the groundwater. The literature review showed different assessments of soil water characteristics and hydraulic conductivity values, which in some cases is difficult to determine through experiment. For near field models, infiltration of water into the containment system and into the waste region is a principal way of inducing the release of radionuclides or chemicals from a containment system. Because water flow is a complex process for the variability of the soil properties, nature of rainfalls, simplifications needs to be made in the performance assessment in order to estimate the infiltration rate (Yim & Simonson, 2000).

DEVELOPMENT OF THE TRANSPORT MODULE

The pathway of the model is the migration of contaminant groundwater from the containment system by advection, which is the movement of contaminants along with flowing ground water at the seepage velocity in porous medium (meaning water). The mercury move from the containment system downward through the unsaturated zone to the saturated zone (aquifer) In the aquifer the mercury concentration are transported by advection-dispersion, and retardation which is a mixing process caused by velocity variations in the porous medium and then water contaminated is withdrawn from the well.

The contaminant transport module in GOLDSIM will simulate this migration. The contaminant transport module consists of linked different elements in GOLDSIM (a one dimension containment system, and unsaturated and saturated groundwater flow). The modeling domain was revised to include the full extent of the EMWMF in its present location (USDOE, 2010). EMWMF facility has an area of about 107,956 square meters (m^2); Table 3 shows the domain characteristics for the model and the location of the hypothetical receptor.

Table 3 Domain characteristic containment system

Parameter	Value
Length of repository (m)	137
Width of repository (m)	788
Distance to well (hypothetical receptor) – X coordinate (m)	-1
Source: USDOE, 2010.	

1.3 Hydrological cycle

Rainfall and snowmelt, can flow into rivers and streams, return to the atmosphere by evaporation or transpiration, or leak into the ground to become part of the subsurface or underground water. As water percolates down through cracks and pores of soil and rock, it passes through a region called unsaturated zone, which is characterized by the presence of both air and water in the spaces between soils particles. Water in the unsaturated zone, is essentially unavailable for use because it cannot be pumped. In the saturated zone, all spaces between soils particles are filled with water. Water in the saturated zone is called the water table (Masters, 1997).

1.4 Mercury containment system

Containment systems are often constructed to prevent, or significantly reduce, the migration of contaminants in soils or ground water. A containment system is necessary whenever contaminated materials are to be buried or left in place at a site. In general, a containment system is performed when extensive subsurface contamination at a site excludes excavation and removal of wastes because of potential hazards, unrealistic costs, or lack of adequate treatment technologies (Van Deuren et. al., 2002).

This conceptual design includes a perimeter dike; a natural or constructed underlying geologic buffer (clay liner) up to 10 ft thick; a 6-ft multilayer base liner system consisting of man-made and natural materials, double leachate collection and detection systems, and a protective soil layer; and a 16-ft multilayer cell cover. An on-site alternative conceptual cross section of the disposal cell is show in Figure 5. The perimeter dike provides stability and guards against erosion. The geologic buffer and multilayer base system reduces the potential for contaminants leaching into the groundwater. The permanent cover minimizes liquid penetration into the closed disposal cell over the long term promotes drainage and minimizes erosion or abrasion of the cover, accommodates settling and subsidence to maintain the integrity of the cell cover, discourages intrusion of humans, animals, and plants, and minimizes maintenance requirements.

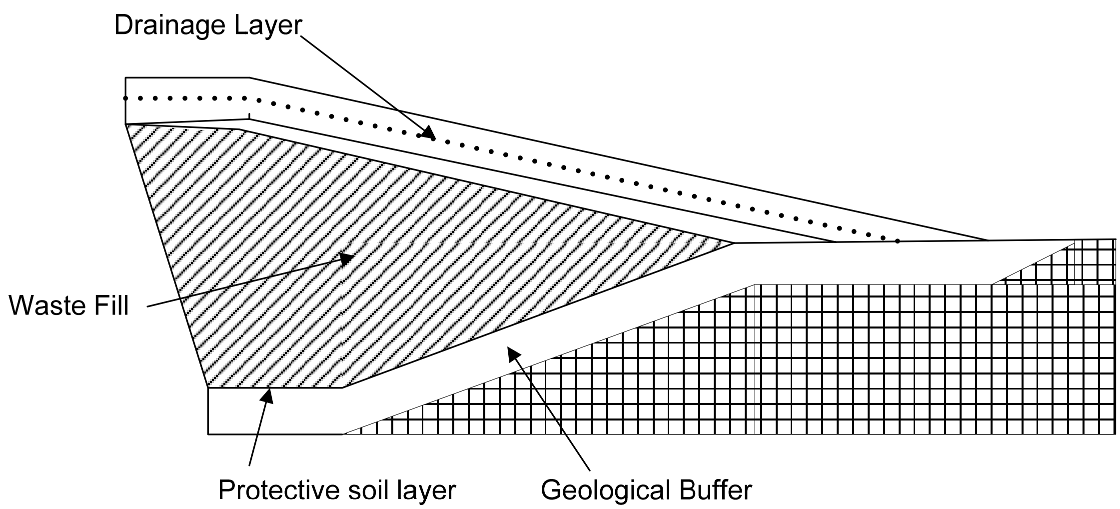

Figure 5 On-site alternative conceptual cross section of disposal cell

Beginning with preliminary design, contingencies will be made that will address shallow groundwater collection and treatment in the event that compliance is ever generated. The final design and size of the cell will depend on the actual amount of waste anticipated, additional information on the geotechnical aspects of the site, and the final waste forms to be disposed. While components may differ from the FS conceptual design, cell performance will not be compromised (USEPA, 2000).

The EMWMF is located at 35° 58’ 13.32” North and 84° 17’ 23.89’’ West with an elevation of 1017 ft. Figure 6 shows the location of the mercury containment system and the drinking water well GW-904 for the hypothetical receptor with a total area of 107956 m^2.

Figure 6 Aerial photo of the mercury containment system at EMWMF

1.5 Develop a contaminant transport module in GOLDSIM

The GOLDSIM Contaminant Transport (CT) module is a mass transport model, which is a mathematical representation of an actual system (GOLDSIM User's Guide (V10.11), 2010). The GOLDSIM contaminant transport module allows one to simulate the transport of mass through an environmental system by providing a number of specialized GOLDSIM elements. The most important of these specialized elements are the transport pathways. Transport pathways represent physical components through which a contaminant species can move, or be stored, such as soil compartments. The general properties of the transport pathways and the environmental medium are defined. To create an environmental system is done by defining a network of transport pathways. A network is a connection of individual pathway transport via mass flux links, a mass flux links defines the mechanisms by which species move between pathways.

For the mercury containment system, the advective mass flux link was used. In an advective mass flux link, a quantity of a medium is specified to flow from one pathway to another, carrying dissolved, sorbed or suspended species with it.

Based on the properties of each pathway, the medium in each pathway, the species, and the specified mass flux links, GOLDSIM contaminant transport module computes the temporally varying concentrations of each pathway's medium, as well as the mass fluxes between pathways. Therefore, the fundamental output of a pathway element is a series of vectors, the mass of the species in each pathway, the concentration of each species within each environmental medium in the pathway, and the mass flux of each species to each of the pathways to which it is connected via mass flux links.

The objective of many contaminant transport studies is to compute contaminant concentrations or flux rates at various locations in the environment and the impact of these contaminants on specific receptors.

GOLDSIM allows defining specific receptor, and associating these with various pathways in the environmental system. The total impact to a receptor is then computed as the sum of the impacts associated with each pathway through which the receptor is exposed to the contaminant. The contaminant transport module for the mercury containment system is constructed, by creating, connecting, and manipulating different graphical objects, which are referred as elements, which represent different components of the model. These elements are the basic for building a model and most elements accept at least one input and produce one or more outputs. There are two main types of elements data and stochastic. A model in GOLDSIM is creating linking the inputs and outputs of one or more elements to other elements. A complex model can have hundreds or thousands of elements and links. GOLDSIM provides a wide variety of built-in elements for entering data and manipulating variables. Each element represents a building block of the model, and has a default symbol by which it is represented in the browsers and menus. The basic GOLDSIM elements can be divided into categories such as input, stock, functions, event, delay, and results. Table 4 provides a description of the capabilities, and limitations of the inputs elements used in the contaminant transport module in GOLDSIM with a brief explanation of the inputs elements used in the module for the mercury containment system.

Table 4 Description of the elements used in the module

Element	Description
Species	This element is used to define a vector containing chemical species (mercury) tracked throughout the model. In most contaminant transport and risk assessment applications, these are considered contaminants Species are stored in this element and is used to define a vector containing chemical species tracked through the model. In contaminant transport and risk assessment applications, are considered to be contaminants

Fluid material	The fluid element is used to specify a fluid material used in the model. The reference fluid, water exists in all parts of the model. Is used to specify a fluid material used in the model. The reference fluid, water, exists in all parts of the model
Data	Store primary data values, and are limited to be deterministic value for single datum or a vector of matrix data for example length, area of the containment system
Stochastic	Like the data element, also store primary data, but assign to the datum a distribution of the modeler's choice. The distribution in sampled from in the execution of multiple realizations of the model. Examples of stochastic element are the PDF distributions for the dry bulk density, porosity, infiltration rate, and partition coefficient.
Container	The container element is primary an organizational tool, and contains other elements or collections of elements, or even other containers. The contents of each may be accessible or inaccessible to other parts of the model. Example is transport, material, dimension, results, and risk. Which contain other elements or groups of elements or even other containers. The contents of each container can be accessible or inaccessible to other parts of the model.
Expression	The expression elements contain a mathematical expression or function, referencing other elements of the model. This expression can operate on scalars, vectors, or matrices level. Example of expression are the dose formula.
Time History	A result element is a convenient way to store the time history of any value result (scalar, vector, or matrix), to display graphically or in a tabular format. Example of time story is the mercury concentration, dose, and risk.
Multi Variate	Is a result element that allows analyzing multiple outputs in graphical or tabular form. They are only available if multiple realizations have been run.
Cell Pathway	Is a principal element for contaminant transport modeling, and is mathematical equivalent of a mixing cell. Cell pathway element are connected to other transport elements (Such as pipes pathway) to create pathways for the movement of material and contaminants An example of cell pathway element is the mercury containment system.
Pipe Pathway	The pipe transport element also provides for the movement of fluids and contaminants in the model via connections to other elements. The pipe can simulate vertical transport in unsaturated zone or horizontal transport in aquifer. example of pipe element are the unsaturated and saturated zone
Solid Material	Any number of solids may be defined for the model using this solid element. These may include rocks, soils, plant biomass, or other solid materials relevant to contaminant transport or risk. Examples are the solid element for the source soil. unsaturated and saturated zone
Source: GOLDSIM, 2010.	

For the mercury, containment system a one-dimensional transport model was implemented within GOLDSIM CT module using different transport pathways elements. This basic implementation was done in three parts: mercury containment system or source, which was represent with the cell transport pathways, for both the unsaturated zone transport (UZ) and the saturated zone transport (SZ) a pipe transport pathway was used for each of them. Each of these transport pathways are briefly discussed below.

A cell transport pathway is mathematically equivalent to a mixing cell, and can represent the processes of partitioning, solubility constraint, and mass transport. For the mercury containment system the contaminant mass was assumed to be instantaneously, completed mixed and equilibrated throughout the cell, and the mercury are partitioned between the soil and water based on the partition coefficient and mass volumes of the soil and water. A solubility limit of 1.47 mol/L (Clever et., al 1985) was considered for the solubility constraints for mercury hg(II). The mass transport was defined to be advective mass flux. The pipe transport pathways represent a feature, which behave as a fluid conduit. Mass enters at one end of the conduit, advects through and disperses within the conduit, and then exists at the other end of the conduit. Pipe transport pathways used Laplace transform approach to provide analytical solutions to advectively dominated transport problems involving advection, dispersion, and retardation. The contaminant retardation process represented in the pipe transport pathway was the equilibrium partitioning between water in the pathway and the infill medium in the unsaturated zone and saturated zone areas.

1.6 Module structure

The GOLDSIM contaminant transport module contains 50 input parameters in the contaminant transport module. Among those, 50 parameters are treated as deterministic and seven are treated as stochastic variable. Deterministic parameters usually have less or no variability and can be defined in a single value. Probabilistic parameters are normally associated with much uncertainty and are defined as a probability distribution.

In order to organize, manage, and view the model, the elements are organized into several different levels of subgroups and containers in a hierarchical top-down order. This method allows for detailed exploration of the module. The GOLDSIM contaminant transport module contains five top-level subgroups: material, transport, results, dimensions, and risk; each of which consists of several containers. Figure 7 shows the containers which represent the entire model.

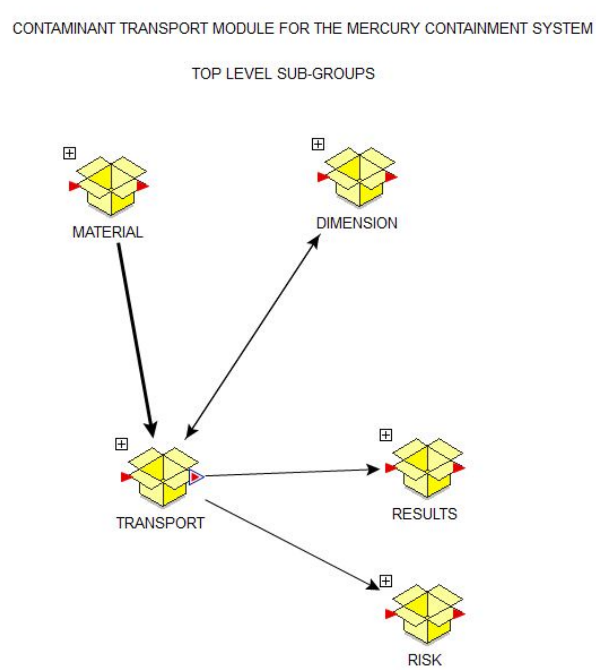

Figure 7 Sub-groups contaminant transport module

The transport subgroup includes 21 input parameters and contains 21 GOLDSIM elements. Based on the characteristics of the input parameters and what they represent, the parameters are grouped into four containers: source, unsatured zone (UZ), saturated zone (SZ), and well. These containers are interconnected to establish the contaminant transport module. The source container stores the input parameters related to the mercury containment system such as the initial concentration, the infiltration rate, etc. Table 5 listed the input parameters used in the source container which are the values used for the calculation package for the analysis of performance of cells 1-6 report (USDOE, 2010). The building structures of the source container are show in Figure 8.

Table 5 List of input parameters used in the source container

Input	Value	Units	Description	Reference
Porosity	0.25		Parameter soil properties element	a
Tortuosity	1		Parameter soil properties element	b
Kd	500	ml/g	Parameter soil properties element	c
Water	1.67×10^{6}	m^3	Medium Total volume	a
Source_soil	2.672×10^{9}	kg	Dry bulk *total waste volume	a
Initial inventory	26720000	g	Water content waste* total waste volume	a
Flow rate (outflow) cell	9.82×10^{2}	m^3/yr	Infiltration rate* Area disposal cell	a
Infiltration rate	9.10×10^{-3}	m/yr	Deterministic value	a
Data from: a (USDOE, 2010) b (GOLDSIM, 2010) c (Katsenovitch, 2009)				

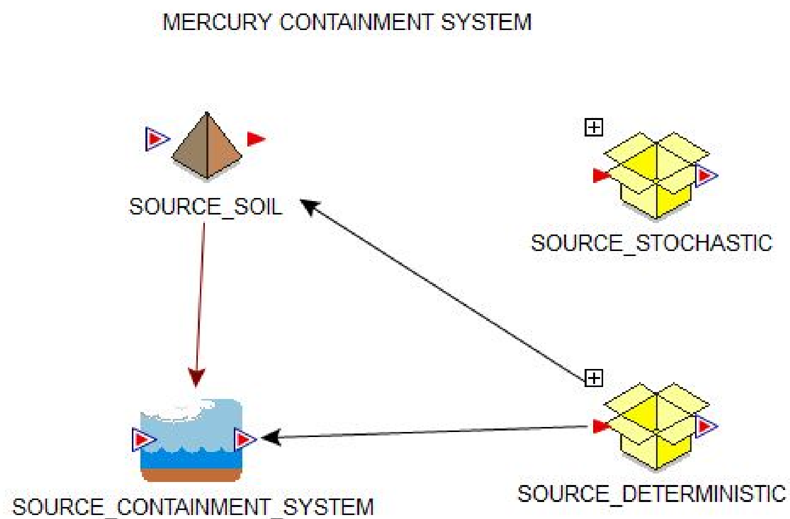

Figure 8 Building structure for source container

The transport container is show in Figure 9. The UZ container and the SZ include the partition coefficient, porosity, dry bulk density, among others. Table 6 lists the parameters used in the unsaturated zone (UZ) and Table 7 lists the parameters used in the saturated zone (SZ) container. Both containers use values for the calculation package for the analysis of performance of cells 1-6 report (USDOE, 2010). The building structures of these parameters for the unsaturated zone are shown in Figure 10 and Figure 11.

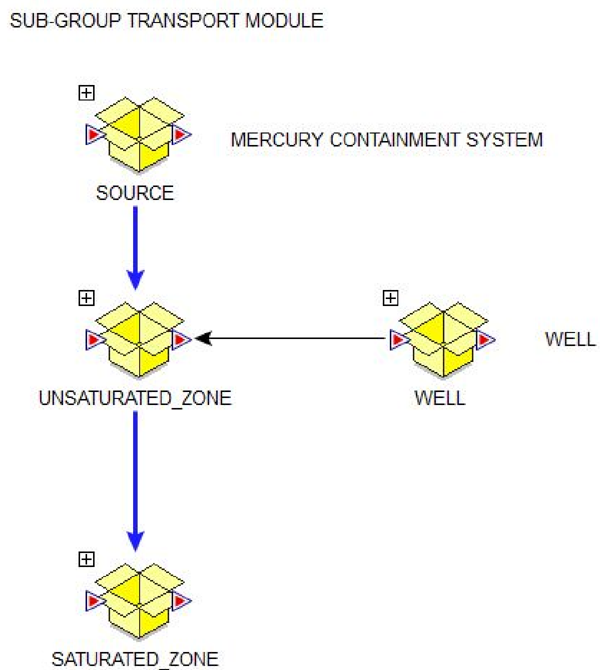

Figure 9 Sub-group transport module

Table 6 List parameters used in the unsaturated zone

Input	Value	Units	Description	Reference
Length	15	m	Thickness of unsaturated zone	a
Area	107956	m^2	Model area (Length and wide)	a
Perimeter	1314.26	m	4*sqrt(model area)	b
Dispersivity	6	m	Longitudinal dispersivity	a
Infill medium	--	--	Unsaturated zone solid properties	a
Fluid Saturation	1	--	Default value	b
Source zone length	137	m	Length disposal	a
Flow rate (inflows) pipe		m^3/yr	(infiltration rate*Area disposal)	a
Flow rate (outflows) pipe	1	m^3/yr	(hydraulic conductivity)*(Area of containment system)	a

Input	Value	Units	Description	Reference
Dry bulk density	1600	kg/m^3	Parameter soil properties element	a
Porosity	0.25	--	Parameter soil properties element	a
Tortuosity	1	--	Parameter soil properties element	b
Partition coefficient	41	ml/g	Parameter soil properties element	c
Flow rate (inflows) Cell	1	m^3/yr		b
Data from: a (USDOE, 2010) b (GOLDSIM, 2010) c (Katsenovitch, 2009)				

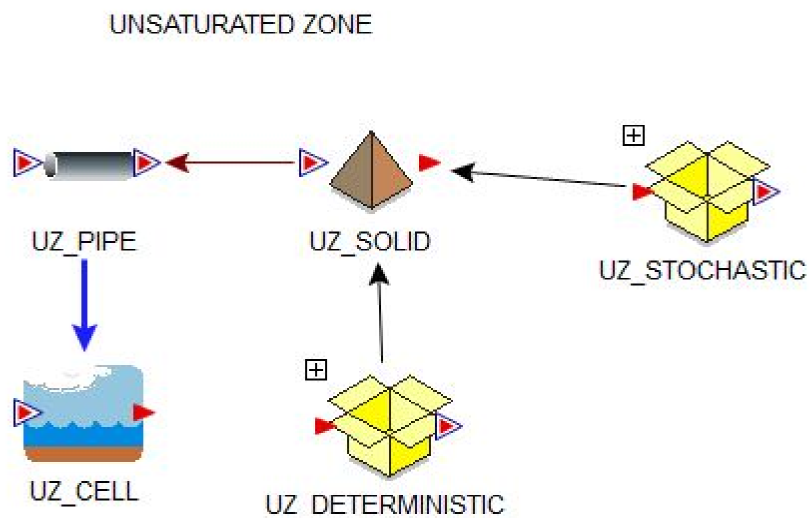

Figure 10 Building structure for unsaturated zone

Table 7 List of parameters used in the saturated zone

Input	Value	Units	Description	Reference
Length	1	m	Distance to well	a
Area	9456	m^2	Thickness SZ*width disposal*2	a
Perimeter	1314.26	m	4*sqrt(model area)	a
Dispersivity	6	m	Longitudinal dispersivity	a
Infill medium	--	--	SZ solid properties	
Fluid saturation	1	--	Default value	b
Source zone length	0	m		b
Input rate		g/yr	Output UZ pipe	b
Flow rate (outflows) pipe	1	m^3/yr	Depends of hydraulic conductivity and area containment	a
Flow rate (inflows) Cell	457733.4	m^3/yr	Horizontal aquifer* area disposal	a
Dry bulk density	1800	kg/m^3	Deterministic value	a
Porosity	0.04		Parameter soil properties element	a
Tortuosity	1		Parameter soil properties element	b
Partition coefficient	100	ml/g	Parameter soil properties element	c
Flow rate (inflows) Cell	1	m^3/yr		a
Data from: a (USDOE, 2010) b (GOLDSIM, 2010) c (Katsenovitch, 2009)				

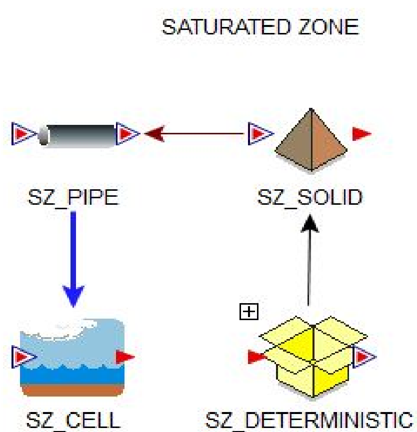

Figure 11 Building structure of the saturated zone

The transport geometry of the saturated zone, which is represented by the pipe pathway element, is shown in Figure 12. The output from the unsaturated zone, which is also represented by a pipe pathway element, is the mass input to the saturated zone (mg/yr). The mass input joins the Q of water (flow rate) in the aquifer in the direction of the drinking water well GW-904.

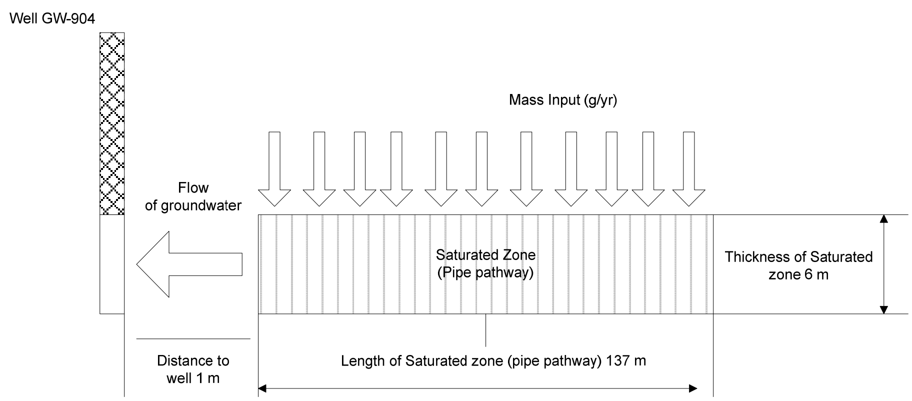

Figure 12 Transport geometry of the saturated zone to the receptor

The dimension container includes all the physical values of the mercury containment system such as the area of the containment system, thickness of the saturated zone, and length of the containment system. Table 8 lists the parameters of the dimension container.

Table 8 List of input parameters used in the dimensions container

Input	Value	Units	Description	Reference
Width disposal waste	788	m		a
Length disposal	137	m		a
Area disposal	107956	m^2	Length*width	a
Total waste volume	$1.67x10^6$	m^3		a
Initial concentration of mercury Hg(II)	1	mg/kg		a
Concentration mercury in the containment system	$1.67x10^6$	mg	Mercury concentration*total waste volume*dry bulk density	a
Length well casing	10	m	Water content waste* total waste volume	a
Longitudinal dispersivity	6	m		a
Distance to well	1	m		a
Stream flow rate	1	m^3/yr		a
Diameter well	1	m		a
Horizontal aquifer velocity	4.24	m/yr		a
Thickness UZ	15	m		a
Thickness SZ	6	m		a
Waste water content	1	--		a
Data from: a (USDOE, 2010)				

The results container includes the results elements time-history, multi-variable statistical analysis, distribution, for the concentration of mercury are listed in

Table 9 for the saturated zone.

Table 9 Output result for concentration of mercury

Output	Units	Description
Concentration in water	mg/L	Time-history
Result distribution	mg/L	Probability of not exceeding
Multi-Variable	mg/L	Sensitivity analysis

For the material container, the inputs elements such as molecular weight 271.50 g/mol and the other elements are used their default value, all of which are listed in Table 10.

Table 10 Input parameters for the material container

Input	Value	Units	Description	Reference
Molecular weight	271.50	g/mol	Value for mercury Hg(II) in element species	a
Reference Diffusivity	1x10-9	m^2/s	Default value	b
Diff. Reduction Formula	1	--	Default value	b
Relative diffusivities		1	Default value	b
Solubility mercury Hg(II)	1.47	Mol/l		a
Data from: a (GOLDSIM, 2010) b (Clever et. al., 1985)				

The risk container includes the calculation of the dose and the risk of the mercury over time (time history). The time history is covered in the risk assessment model.

1.7 Limitation and consideration of the module

The Contaminant Transport Module is a mass transport model, not a flow model. That is, it does not directly solve for the movement of medium through the environmental system being modeled. Therefore, it must directly enter the medium flow rates associated with an advective flux link or provide GOLDSIM with the equations for computing them. The pipe pathway element capacities can provide an exact solution to a very complex physical system. The limitations of the pipe pathway are that they cannot apply solubility limits within the pathway, species are discharged from a pathway based on the properties of the pathway at the time the species entered it, and lopping reaction chains are not permitted. Properties of the cell pathway element are flexibility, stability, and accuracy; but it is tedious to construct networks and numerical dispersion.

DEVELOPMENT OF THE RISK ASSESSMENT MODEL

The purpose of the probabilistic risk assessment model for simulations applying to the mercury containment system is to provide information about the effect of selected stochastic parameters on the dose and risk for mercury existence in the drinking water obtained from well GW-904 located near the EMWMF and Bear Creek. A probabilistic analysis is a statistical technique for studying the expected behavior of a system with parameters whose values are uncertain. The simulation consists of hundreds or thousands of deterministic Monte Carlo realizations.

In GOLDSIM the probabilistic distributions for the selected input parameters are to be treated as stochastic parameters, such as infiltration rate, porosity, partition coefficient, and dry bulk density; while the rest of the parameters stay as a deterministic value. The Monte Carlo analysis includes the total simulation, time duration, and the total realization number for the probabilistic simulation. All these parameters are represented by a probability density function (PDF). The outputs from the probabilistic simulation model, such as the mercury concentration, dose, and risk, are also PDF distributions.

An important step in Monte Carlo Analysis (MCA) is the selection of the most appropriate probability distribution functions to represent the parameter to evaluate if they have a strong influence on the concentration of contaminants, and therefore an influence on the dose and risk estimates. In a probabilistic risk assessment (PRA) a PDF, also referred to as a probability model, characterizes the probability of each value occurring from a range of possible values (USEPA, 2001). One advantage of using a PDF is that its distribution represents a large set of data values in a compact way (Law & Kelton, 1991). Developing site-specific PDFs for selected input variables or toxicity values can be time and resource intensive, and in many cases, may not add value to the risk management decision. Table 11 lists some examples of probability distributions commonly used in PRAs with their theoretical bounds and parameters values. Many of these distributions given in Table 11 can assume flexible shapes; they offer practical choices for characterizing variability (USEPA, 2001).

Table 11 Theoretical bounds and parameters for selected distributions for PRA

Distribution	Parameters	Theoretical Bounds
Normal	(μ,σ)	$(-\infty, +\infty)$
Lognormal	(μ,σ)	$[0, +\infty)$
Weibull	(α,β)	$[0, +\infty)$
Exponential	(β)	$[0, +\infty)$
Gamma	(α, β)	$[0, +\infty)$
Beta	$(\alpha1,\alpha2, a, b)$	[a, b]
Uniform	(a, b)	[a, b]
Triangular	(a,m, b)	[a,b]
Empirical (Bounded EDF)	(a,b, {x}, {p})	[a,b]

[1]a=minimum b=maximum, μ =mean, σ=standard deviation, m=mode, α=shape parameter, x=value, p=probability. Source: USEPA, 2001

It is generally assumed that a hydrological variable has a certain distribution type. Most of the common and important probability distributions used in hydrology are the normal, lognormal, Gamma, Gumbel, and Weibull (Aksoy, 2000). The normal and lognormal are generally used to fit annual flows in rivers. The Gamma distribution has the advantage of having only positive values since hydrological variables, such as rainfall, infiltration rate, and runoff, are always positive and greater than zero or equal to zero at a lower limit value (Markovic, 1965). These three distributions were selected to create the PDF distributions for the parameters porosity, dry bulk density, infiltration rate, and partition coefficient after a selection process.

Recent developments in hydrological modeling, flood risk analysis, and environmental impact assessments have demonstrated the usefulness of random variable simulations (NRC, 2000). An EPA (1992) report recommend that where there is a question about the distribution of the data set, a statistical test should be used to identify the best distributional assumption for the data set.

1.8 Dose and risk calculations in the model

Dose-response is performed with the main objective of obtaining a mathematical relationship between the amount of a contaminant that a human is exposed to and the risk that there will be an unhealthy response to that dose (Masters, 1997).

For the residential receptor exposure, the ingestion of chemicals in drinking water and other beverages containing drinking water is calculated using Equation 2 (USEPA, 1989).

$$Intake = \frac{[(CW)(IR)(EF)(ED)]}{[(BW)(AT)]} \tag{2}$$

Where Intake is mg/kg-days, CW is the chemical concentration in water (mg/L), IR is the ingestion rate (liters/day), EF is the exposure frequency (days/year), ED is the exposure duration (years), BW is the body weight (kg), and AT is the averaging time, which is the period over which exposure is averaged (days). For non-carcinogenic effects, the intake becomes Equation 3.

$$Intake = \frac{[(CW)(IR)]}{[(BW)]} \tag{3}$$

In which IR, the ingestion rate, is 2 liters per day (adult, 90^{th} percentile, EPA 1989d) and BW is 70 kg (adult, average; EPA 1989d). Equation 4 is used to calculate the dose of mercury in contaminated groundwater extracted from the well.

$$Dose = (CW)(IR)(365\frac{days}{years}) \tag{4}$$

Where dose is mg/yr and 365 days/year is a conversion factor. Equation 5 for risk is as follows:

$$Risk = \frac{(Intake)}{(Rfd)} \tag{5}$$

Where risk is dimensionless, intake is mg/kg-days, and Rfd is 0.0003 (mg/kg/day).

Equation 5 was used to evaluate human exposure to the contaminated groundwater from the well. The dose and risk assessment was computed based on contaminant concentrations of mercury in the groundwater. The equations and methodologies used are typical of those recommended by the EPA.

The projected risk and doses of mercury for a period of 10,000 years after closure of the containment system were calculated for consumption of drinking water from a well. Table 12 shows the drinking water standards, regulations, and health advisories for inorganic mercury and Table 13 shows the oral reference dose (RfD) and the Drinking Water Equivalent Level (DWEL), defined by the EPA as “a lifetime exposure concentration protective of adverse, non-cancer health effects, that assumes all of the exposure to a contaminant is from drinking water.”

Table 12 Drinking water standards and health advisories for mercury

Chemical	CASRN	Standard Status Reg.	Standard MCLG (mg/L)	Standard MCL (mg/L)
Inorganic mercury	7487-94-7	F	0.002	0.002
Data from: USEPA, 2004				

Table 13 Oral reference dose and drinking water equivalent level for mercury

Chemical	RfD (mg/kg/day)	DWEL	Lifetime (mg/L)	mg/L at 10^{-4} Cancer risk	Cancer group
Inorganic mercury	0.0003	0.01	0.002	----	D
Data from: USEPA, 2004					

In Table 12, the MCLG is the Maximum Contaminant Level Goal, a non-enforceable health goal which is set at a level at which no known or anticipated adverse effect on the health of a person occurs and which allows an adequate margin of safety. The MCL, or Maximum Contaminant Level, is the highest level allowed of a contaminant in drinking water. MCLs are enforceable standards that are set as close to the MCLG as feasible using the best available analytical and treatment technologies and considering costs. RfD is the reference dose, which is an estimate of a daily oral exposure to the human population that is likely to be without an appreciable risk of deleterious effects during a lifetime. According to the USEPA 1986 guidelines, which established a qualitative weight of evidence judgment as to the likehood that a chemical may be a carcinogen, the inorganic mercury belongs to group D, indicating evidence of no carcinogenicity for humans.

According to table 1200-5-1-0.9(I) (d) in chapter 1200-5-1-09 of the Detection Limits for Inorganic Contaminants Rules of the Tennessee Department of Environment and Conservation (TDEC), the maximum concentration level for inorganic mercury permitted in the water of the public system is 0.002 mg/L (TDEC, 2006). The building structure for the risk container, as shown in Figure 13, store expression elements, such as dose and water intakes.

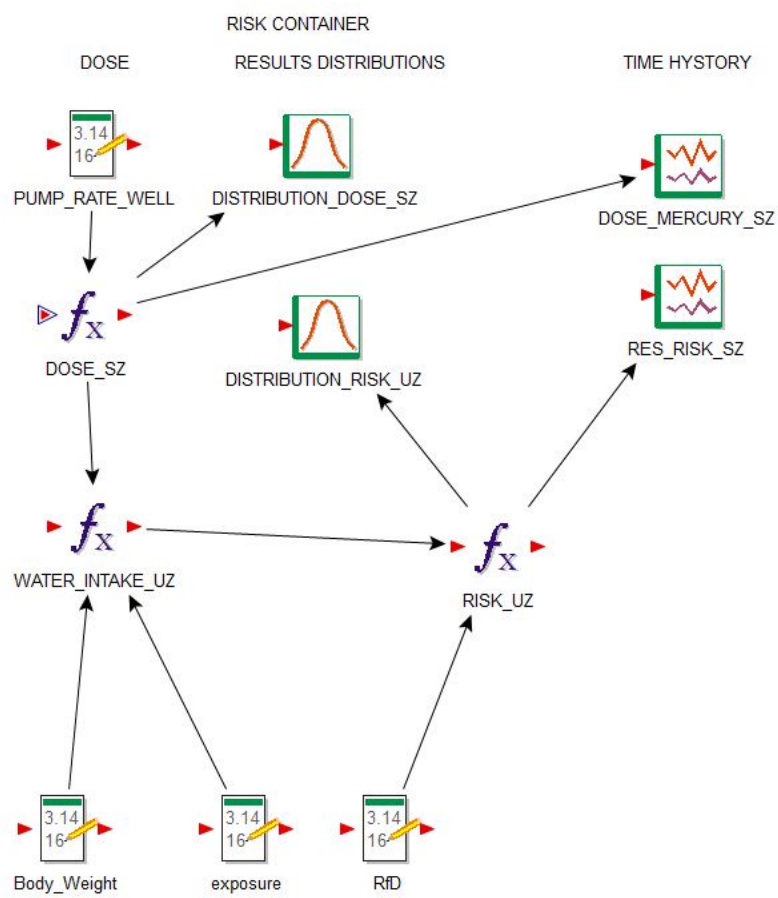

Figure 13 Building structure risk container

1.9 Mercury and their MCLs

National Primary Drinking Water Regulations (NPDWRS), or primary standards, are legally enforceable standards that apply to public water systems. Primary standards protect public health by limiting the levels of contaminants in drinking water. The maximum contaminant level (MCL) for inorganic mercury is 0.002 mg/L and the potential health effects from long-term exposure above the MCL is kidney damage. The sources of contamination in drinking water are erosion of natural deposits, discharge from refineries, factories, and runoff from landfills and croplands (USEPA, 2009).

1.10 Uncertainty in the risk assessment model

Uncertainty analyses and probabilistic approaches have also been used for decommissioning of contaminated sites. Sites contaminated by hazardous materials generally show high degrees of variability in concentrations of contaminants and in natural environmental characteristics that affect transport. The word "variability" is issued to describe the fact that the actual characteristics exist in different values at different points in space and time. This has been termed "natural or intrinsic uncertainty" by others (Benjamin and Cornell 1970; Vicens et al., 1975). These sites can also have substantial levels of uncertainty in these parameters, in that any measurement of them has some degree of error. The term "uncertainty" is used to describe the fact that the exact values of the variables are not known, but are estimated by limited measurements. Therefore, stochastic analysis techniques that explicitly consider site variability and uncertainty would be more appropriate for use at these sites (Batchelor et. al., 1998).

1.11 Selection and fitting of distributions

U.S. Nuclear Regulatory Commissions pay special attention to the decommissioning process because of elevated levels of radioactive contaminants. Decisions about the safety of the Site Decommission Management Plan (SDMP) sites are likely to be made in an atmosphere of significant uncertainty, arising from a number of conditions. These conditions include the presence of long-lived radionuclides requiring exposure predictions 1,000 years or more into the future, potential exposure through multiple pathways, limited data characterizing the hydrological performance of the subsurface, and simplifications to the physical system and the transport mechanisms to reduce the computational requirements of the exposure analysis.

Because site-specific data on the soil hydraulic parameters used in these programs are often not available, NRC must make assumptions regarding the parameter values to use to estimate dose impacts from SDMP sites. Generic probability distributions, such as normal, lognormal, and Beta for unsaturated and saturated zone soil hydraulic parameters, are presented. These generic distributions are compared to the default or recommend parameter values from other sources. The generic distributions are useful for modeling the uncertainty in soil hydraulic parameters when information about the soils at a site is limited to the soil texture (NRC, 1997).

The choice of a distribution type is based on professional judgment and has greater uncertainty when data is insufficient. The normal distribution is suggested for use as subjective probability distributions in analysis of additive models (DOE 1994). The determination of a data distribution is accomplished by following Environmental Protection Agency guidance for data quality assessments, which are practical methods for data analysis (EPA, 2000). The normal distribution was chosen based upon EPA CERCLA risk assessment guidance, which prescribes the use of normal or lognormal distributions. For this analysis, a selection of three of five distributions, which are normal, lognormal, Gamma, Beta, and Weibull were used based on a coefficient of variation selection process.

1.12 Monte Carlo method

The selected input parameters are unsure, therefore the prediction of the future performance of the containment system is necessarily uncertain. The results of any analysis is based on inputs represented by probability distributions is itself a probability distribution.

In order to compute the probability distribution of predicted performance, it is necessary to propagate or translate the input uncertainties into uncertainties in the results. One common technique for propagating the uncertainty in the various aspects of a system to the predicted performance, and the one used in GOLDSIM, is a Monte Carlo simulation.

In a Monte Carlo simulation, the entire system is simulated a large number of times. Each simulation is equally likely, and is referred to as a realization of the system. For each realization, all the uncertain parameters are sampled. The system is then simulated through time. The results are a large number of separate and independent results, each representing a possible future for the system. The results of the independent system realizations are assembled into probability distributions of possible outcomes (GOLDSIM, 2010). A schematic of the Monte Carlo method is shown in Figure 14.

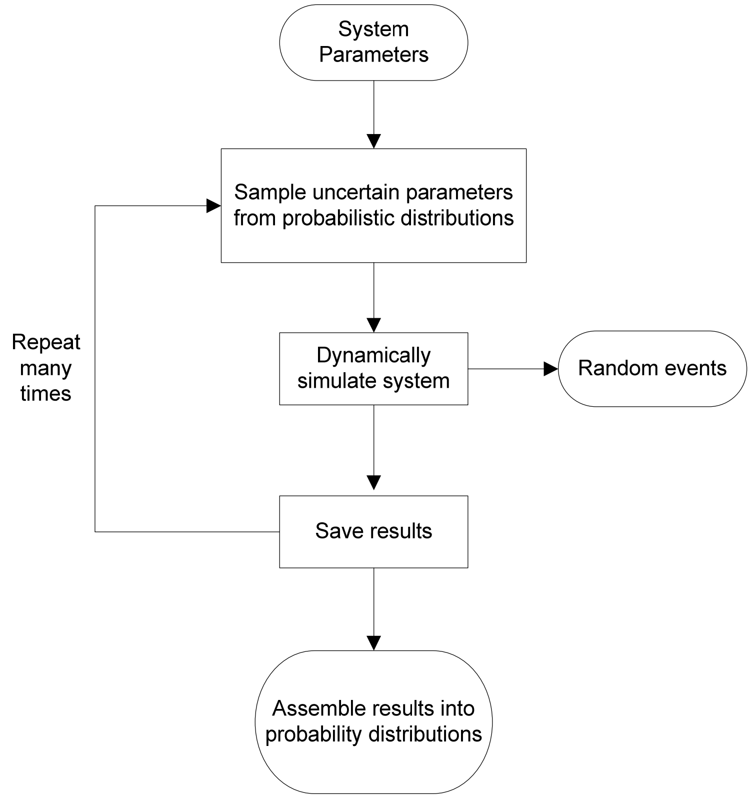

Figure 14 Schematic of a Monte Carlo simulation

In a Monte Carlo method a large number of particles of solute are stochastically followed according to the appropriate probability distribution functions describing their motion and undergone processes (de Marsily, 1986).

1.13 Uncertainty parameters

1.13.1 Infiltration rate

The infiltration rate is the process of vertical movement of water into soil from rainfall. Infiltration of water plays a key role in the transport of chemicals into the subsurface (Bedient et. al., 1994). The single value for the infiltration rate is 0.91 cm/yr from the report Calculation Package for the Analysis of Performance of Cells 1-6, with Underdrain, of the Environmental Management Waste Management Facility, Oak Ridge

TN (USDOE, 2010) for inside the boundary conditions.

In order to have a probabilistic density function (PDF) distribution for the infiltration rate, a set of data was used from the Harden (2003) study. In this study, which is about infiltration on mountains slopes in Oak Ridge Reservation, the values for the infiltration rate were too high to be used for the fitting. Instead, a sample of 50 random numbers was created in Excel, which are listed in Table 54 of the appendix. The distribution fitting tool in Matlab program (Mathworks, 2002) was used to find the best fitting distributions for the infiltration rate. The results are illustrated in**Error! Reference source not found.** Figure 15, which shows the fitting for the normal, lognormal, Gamma, Beta and Weibull distributions for infiltration rate data. Table 14 shows the results of the calculation for each distribution with their mean and standard deviation, which is for the infiltration rate stochastic parameter.

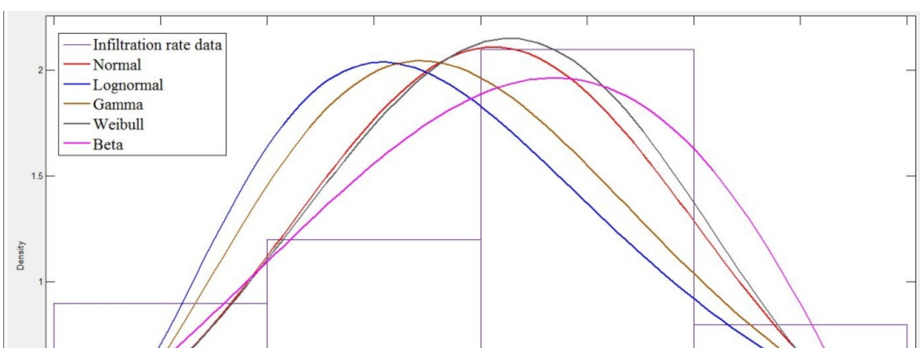

Figure 15 Fitting of PDF distributions for an infiltration rate data

Table 14 Parameters for PDF distributions for infiltration rate data

Distribution	Mean	Variance
Normal	0.61	0.036
Lognormal	0.61	0.05
Gamma	0.61	0.04
Beta	0.61	0.034
Weibull	0.61	0.032

In order to select three of the five PDF distributions to represent the stochastic infiltration rate and the rest of the selected parameters, the coefficient of variation (CV), a measurement of spread that is relative to the magnitude of the variable considered, was used. The CV is often the result of the formula CV= (σ/μ) in which σ is the standard deviation and μ is the mean.

Table 15 Coefficient of variation for an infiltration rate

Distribution	Coefficient of Variation CV (%)
Normal	5.90
Lognormal	8.19
Gamma	6.55
Beta	5.57
Weibull	5.54

A set of deterministic simulations were performed for each of the PDF distributions, with the purpose of evaluating their output, which is the concentration of mercury. Figure 16 shows the 95th percentile for concentration of mercury for deterministic simulations. The most conservative values of concentration are the highest, and based on this graph the distributions normal, lognormal and, Gamma represented the most conservative values of concentration, and were therefore selected for the rest of the parameters. Using the coefficient of variation formula, the normal distribution was selected for the infiltration rate stochastic parameter.

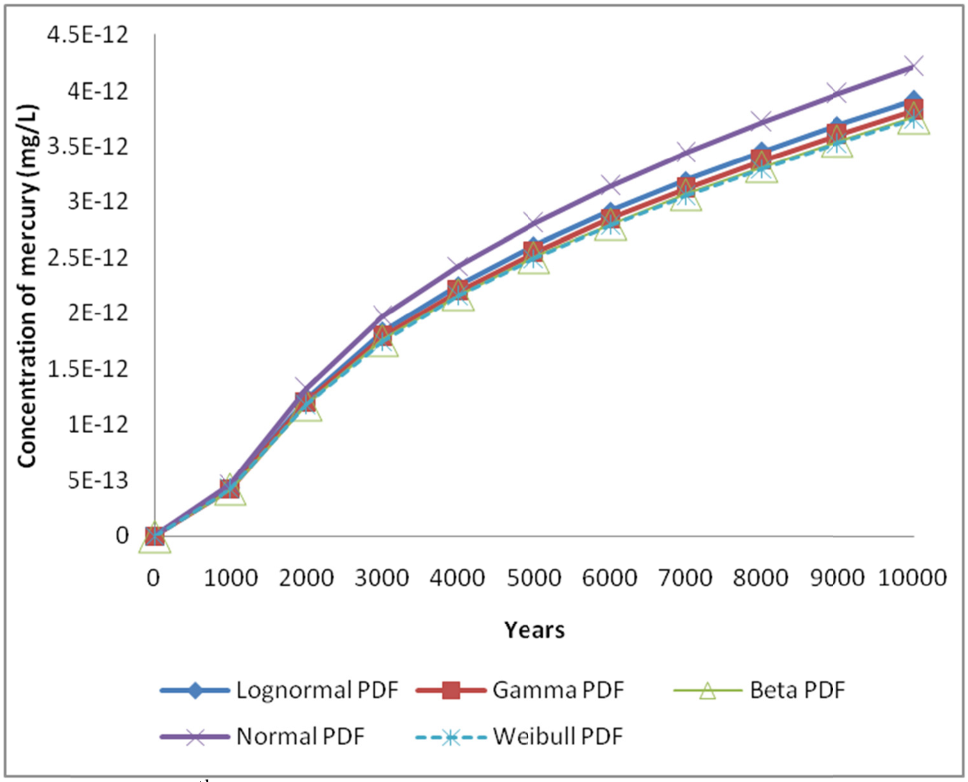

Figure 16 95th concentration of mercury for all PDF distributions for infiltration rate

1.13.2 Dry bulk density

Bulk density is necessary to calculate the retardation factor. The bulk density is equal to the mass of solid divided by the total volume occupied by solid, liquid, and gas. The bulk density is correlated to the effective porosity (Ho et. al., 1999).

For this study the dry bulk density for the unsaturated zone (UZ) and the mercury containment system were the same with a value of 1600 kg/m^3 (1.6 g/cm^3), while the dry bulk density for the saturated zone (SZ) was 1800 kg/m^3 (1.8 g/cm^3). These values were taken from the Calculation Package for the Analysis of Performance of Cells 1-6, with Underdrain, of the EMWMF, Oak Ridge TN report (USDOE, 2010).

Values for the PDF distribution for the dry bulk density for the mercury containment system and for the unsaturated zone were taken from the Harden study (2003). A sample of 24 values for bulk density for the top slope position are listed in

Table 55 in the appendix.

Using the Matlab (Mathworks, 2002) distribution fitting tool to find the best fitting distributions for the dry bulk density, **Error! Reference source not found.**Figure 17 shows the fitting for the normal, lognormal and Gamma distributions for dry bulk density data from the unsaturated zone. Table 16 shows the mean and standard deviation for each distribution for the dry bulk density for the source and unsatured zone (UZ).

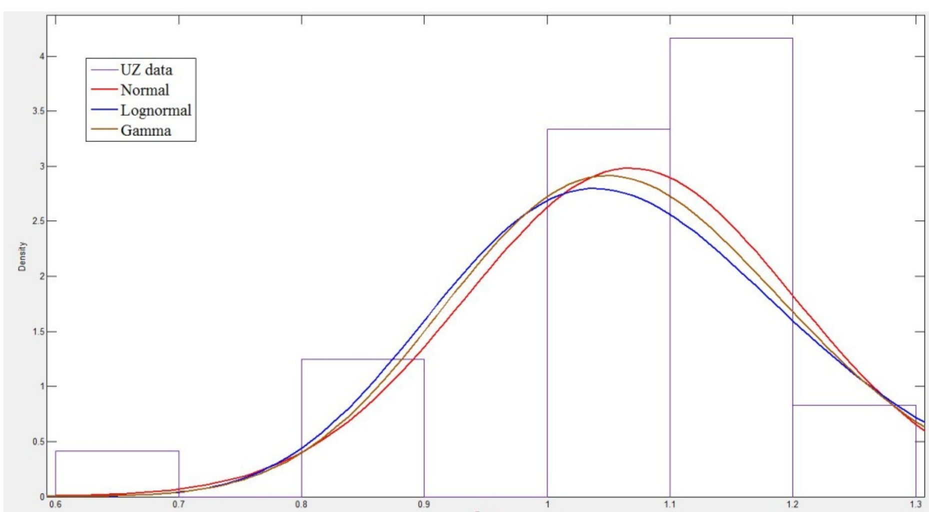

Figure 17 Fitting of PDF distributions for dry bulk density data from the UZ

Table 16 Parameters for PDF distributions for dry bulk density data for UZ

Distribution	Mean	Variance
Normal	1.067	0.017
Lognormal	1.068	0.021
Gamma	1.0675	0.019

In order to select the PDF distribution to represent the stochastic dry bulk density for the source and UZ, the coefficient of variation (CV) was calculated, resulting in the selection of the normal distribution.

Table 17 Coefficient of variation for the dry bulk density data for UZ

Distribution	Coefficient of Variation CV (%)
Normal	1.59
Lognormal	1.96
Gamma	1.77

For the saturated zone, the dry bulk density was selected based on the Harden study (2003), but for this case the values were from the bottom slope position. A sample of 14 values are listed in the Table 56.

Using Matlab (Mathworks, 2002) distribution fitting tool to find the best fitting distributions for the dry bulk density for the saturated zone, Figure 18**Error! Reference source not found.** shows the fitting for the normal, lognormal, and Gamma distribution for dry bulk density data for the saturated zone. Table 18 shows the mean and standard deviation for each distribution for the dry bulk density for the saturated zone (SZ).

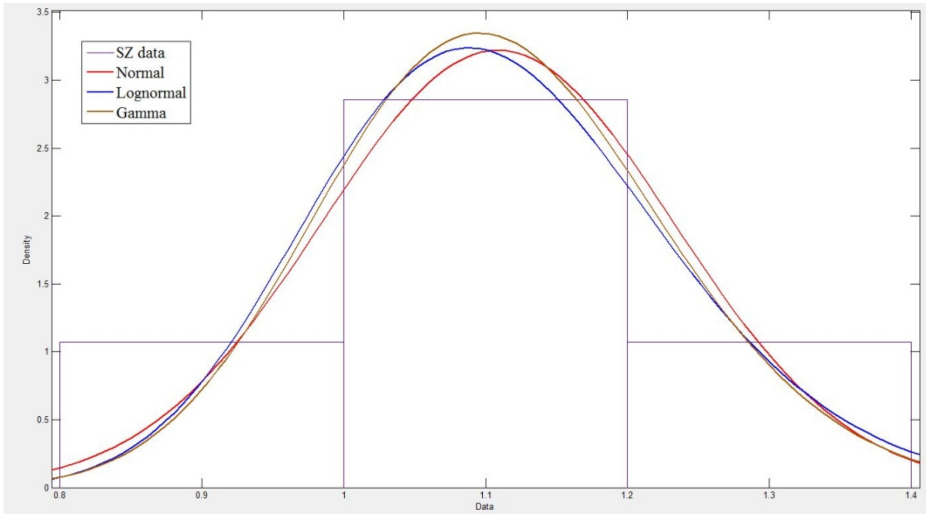

Figure 18 Fittign of PDF distributions for dry bulk density data for SZ

Table 18 Parameters for PDF distributions for dry bulk density data for SZ

Distribution	Mean	Variance
Normal	1.108	0.015
Lognormal	1.109	0.015
Gamma	1.108	0.014

In order to select the PDF distribution to represent the stochastic dry bulk density for the saturated zone (SZ) the CV was calculated, and as a result the gamma distribution was selected.

Table 19 Coefficient of variation for the dry bulk density SZ

Distribution	Coefficient of Variation CV (%)
Normal	1.35
Lognormal	1.35
Gamma	1.26

1.13.3 Porosity

The effective porosity is equal to the volume of pore space that can be occupied by mobile fluid divided by the total volume. If the porosity increases, the retardation factor Rf decreases. It was assumed that the mercury containment system and the unsaturated zone have the same porosity which is 0.25 and for the saturated zone was 0.04. Both values were taken from the Calculation Package for the Analysis of Performance of Cells 1-6, with Underdrain, of the EMWMF, Oak Ridge TN report (USDOE, 2010).

To calculate the PDF distribution for porosity in the unsaturated zone and knowing that porosity can take a positive value between a range of zero and one, a set of 20 random numbers between 0 and 1 was created in Excel, which can be found in Table 57 in the appendix.

Using Matlab (Mathworks, 2002) distribution fitting tool to find the best fitting distributions for the porosity in the unsaturated zone, **Error! Reference source not found.** shows the fitting for the normal, lognormal and Gamma distribution. Table 20 shows the mean and standard deviation for each distribution for the porosity in the source and UZ.

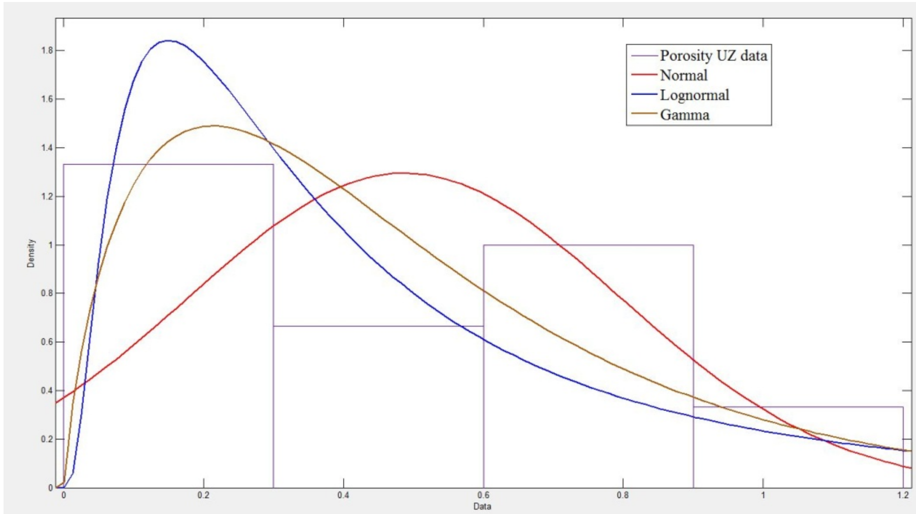

Figure 19 Fitting of PDF distributions and porosity data for UZ

Table 20 Parameters for PDF distributions for porosity data for UZ

Distribution	Mean	Variance
Normal	0.487	0.094
Lognormal	0.554	0.430
Gamma	0.487	0.1333

In order to select the PDF distribution to represent the stochastic porosity for the source and UZ, the CV was calculated, resulting in the selection of the Gamma distribution.

Table 21 Coefficient of variation for the porosity source and UZ

Distribution	Coefficient of Variation CV (%)
Normal	19.30
Lognormal	77.61
Gamma	27.37

The porosity in the saturated zone has a value of 0.04 and knowing that porosity is in the range of 0 and 1, a set of 20 random numbers was created in Excel focusing on a range of 0.01 to 0.25 values. The numbers are listed in Table 58 in the appendix.

The Matlab (Mathworks, 2002) distribution fitting tool was then used to find the best fitting distributions for the porosity in the saturated zone. **Error! Reference source not found.** shows the fitting for the normal, lognormal and Gamma distributions. Table 22 shows the mean and standard deviation for each distribution for the porosity in the SZ.

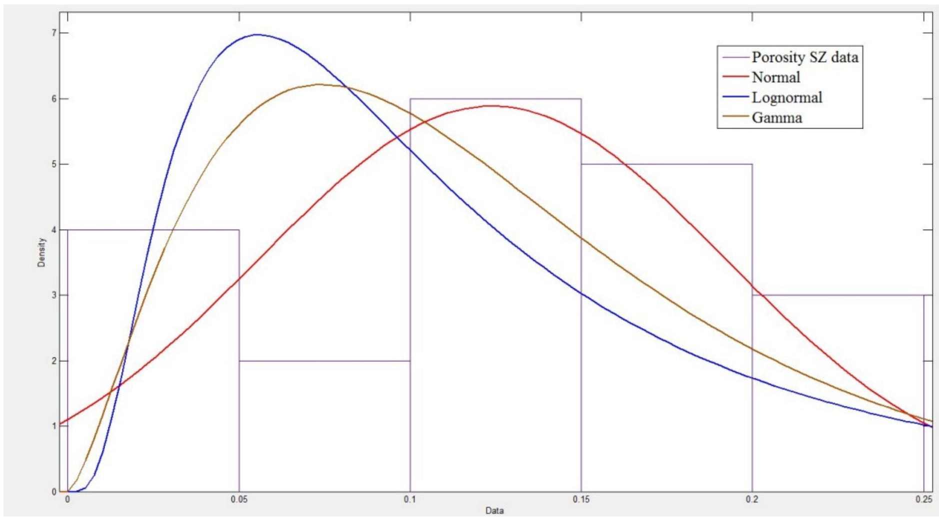

Figure 20 Fitting of PDF distributions and porosity data for SZ

Table 22 Parameters for PDF distributions for porosity SZ

Distribution	Mean	Variance
Normal	0.124	0.004
Lognormal	0.134	0.014
Gamma	0.124	0.006

In order to select the PDF distribution to represent the stochastic porosity for the SZ the CV was calculated. The lognormal distribution was selected instead of the normal distribution, which has the lowest CV for the porosity in the source and UZ, in order to use the widest distribution for more uncertainty.

Table 23 Coefficient of variation for the porosity in the SZ

Distribution	Coefficient of Variation CV (%)
Normal	3.22
Lognormal	10.44
Gamma	4.83

1.13.4 Partition coefficient

The distribution coefficient, Kd, is used to describe the reversible equilibrium partitioning of contaminants between the solid phase and the liquid phase. If the value of Kd is large, then sorption onto the solid phase is large and the retardation factor is large, which reduces the transport quantities of advection and dispersion.

Experimental work using ORR soils finds that the partition coefficient is in the range of 508-511 ml/g (or log Kd of 2.7-2.7 ml/g) (Katsenovitch, 2009). A statistical analysis prepared for the EPA by Allison (2005) estimated a value for soil/water partition coefficient or log Kd, for Hg (II) from 2.2-5.8 ml/g with a mean of 3.6 ml/g. To be conservative, the following values were used for the partition coefficient: 500 ml/g for the mercury containment system, 41 ml/g for the unsaturated zone (DOE, 1994), and 100 ml/g for the saturated zone.

To create the PDF distribution for the partition coefficient for the mercury containment system a sample of 20 random numbers was created in Excel, which are in the range of 200 to 700 ml/g and are listed in Table 59 in the appendix. Using Matlab (Mathworks, 2002) distribution fitting tool to find the best fitting distributions for the infiltration rate, **Error! Reference source not found.**Figure 21 shows the fitting for the normal, lognormal and gamma distributions. Table 24 shows the mean and standard deviation for each distribution for the partition coefficient for the containment.

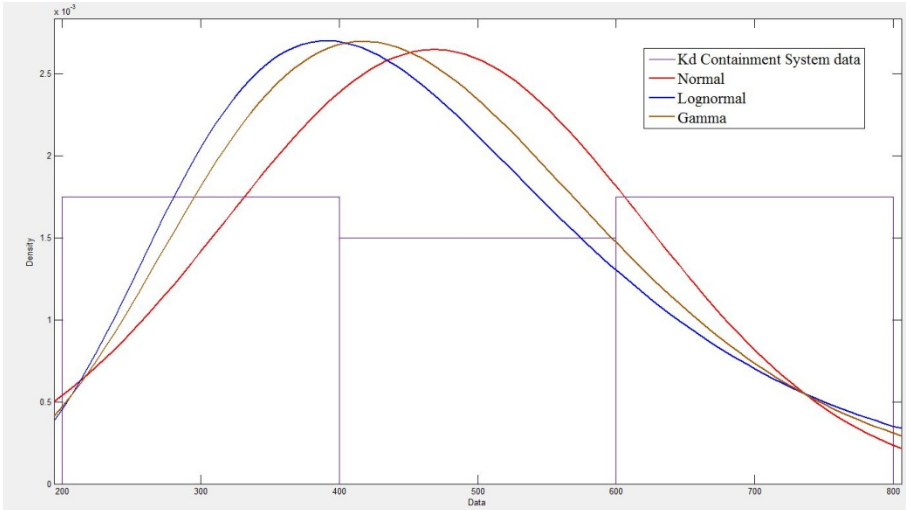

Figure 21 Fittign of PDF distributions and Kd data for containment system

Table 24 Parameters for PDF distributions for Kd containment

Distribution	Mean	Log Mean	Variance	Log Variance
Normal	468	2.67	22700	4.35
Lognormal	472	2.67	29900	4.47
Gamma	468	2.67	24000	4.38

In order to select the PDF distribution to represent the stochastic partition coefficient for the containment, the CV was calculated, and as a result the normal distribution was selected.

Table 25 Coefficient of variation for partition coefficient source

Distribution	Coefficient of Variation CV (%)	Log CV (%)
Normal	4852	163.12
Lognormal	6348	167.41
Gamma	5140	164.06

The partition coefficient for the unsaturated zone is 41 ml/g. In order to select the PDF distribution, a set of 20 random numbers between the values of 20 and 100 was created in Excel, which can be found in Table 60 in the appendix.

Matlab (Mathworks, 2002) distribution fitting tool was used to find the best fitting distributions for the partition coefficient in the saturated zone. **Error! Reference source not found.** shows the fitting for the normal, lognormal, and Gamma distributions. Table 26 shows the mean and standard deviation for each distribution for the partition coefficient for the UZ.

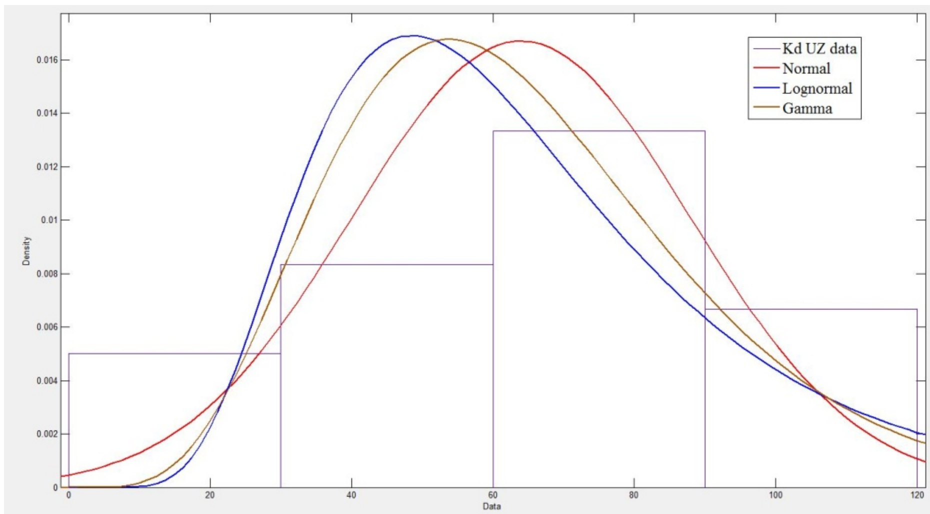

Figure 22 Fitting of PDF distributions for Kd data for UZ

Table 26 Parameters for PDF distributions for Kd unsaturated zone

Distribution	Mean	Log Mean	Variance	Log Variance
Normal	64	1.80	238	2.37
Lognormal	65	1.81	907	2.95
Gamma	64	1.80	652	2.81

In order to select the PDF distribution to represent the stochastic partition coefficient for the UZ, the CV was calculated, resulting in the selection of the lognormal distribution.

Table 27 Coefficient of variation for partition coefficient UZ

Distribution	Coefficient of Variation CV (%)	Log CV (%)
Normal	373	131

Lognormal	1395	164
Gamma	1019	155

The partition coefficient for the saturated zone is 100 ml/g. In order to select a PDF distribution, a set of 20 random numbers was created in Excel focusing on a range of values from 80 to 200. This list can be found in Table 61 of the appendix.

Matlab (Mathworks, 2002) distribution fitting tool was used to find the best fitting distributions for the partition coefficient in the saturated zone. Figure 23 shows the fitting for the normal, lognormal and Gamma distributions. Table 28 shows the mean and standard deviation for each distribution for the partition coefficient for SZ.

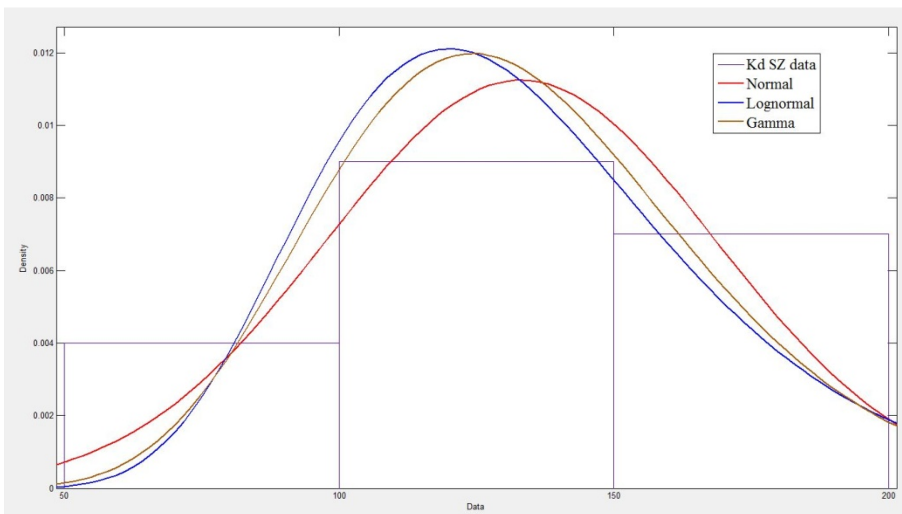

Figure 23 Fitting of PDF distributions for Kd data for SZ

Table 28 Parameters for PDF distributions for Kd data for saturated zone

Distribution	Mean	Log Mean	Variance	Log Variance
Normal	133.1	2.12	1259	3.10
Lognormal	133.32	2.12	1294	3.11
Gamma	133.1	2.12	1173	3.06

In order to select the PDF distribution to represent the stochastic partition coefficient for the SZ, the CV was calculated and the lognormal distribution was selected.

Table 29 Coefficient of variation for partition coefficient data for UZ

Distribution	Coefficient of Variation CV (%)	Log CV (%)
Normal	946	145
Lognormal	970	146
Gamma	881	144

SIMULATIONS OF THE RISK ASSESSMENT MODEL

Defining the different scenarios for the probabilistic risk assessment is vital to be able to evaluate the long-term performance of the containment system. There are a prohibitively large number of scenarios to consider for a 10,000 year period. The key is to identify a manageable set of scenarios that are representative of the conditions most important to reducing the risk and dose of the containment system (Garrick, 2002).

The EPA recommends using the average concentration to represent a reasonable estimate of the concentration likely to be contacted over time (EPA, 1989). The EPA's supplemental guidance to RAGS (USEPA, 1992), explains that because the uncertainty associated with estimating the true average concentration at a site, the 95 percent upper confidence limit (UCL) of the arithmetic mean should be used for this variable.

1.14 Simulations settings

The simulation setting for the GOLDSIM contaminant transport module is the time duration, which for disposal cells containing low-level radioactive wastes are expected to perform for at least 10,000 years (NRC, 2000).

In a Monte Carlo simulation, a single realization represents one possible output using one value of the selected stochastic parameter. A time step is a discrete interval of time used in dynamic simulations (GOLDSIM, 2010).

1.15 Risk evaluation deterministic simulation

The first simulation of the model in GOLDSIM was probabilistic but with all the inputs parameters set as single values (deterministic). The objective of these simulations was to evaluate the output with diferent realization settings. This was done for 10,000 years, using 1000 and 100 realizations. Table 62 and Table 63 in the appendix list the calculations done by GOLDSIM for the mercury concentration, for the two realizations. Figure 24 shows the outputs from GOLDSIM of a probabilistic simulation with all the inputs parameters set as a deterministic value for 1000 realizations where the 95th percentile for concentration of mercury was 5.14×10^{-12} mg/L for the time period of 10,000 years.

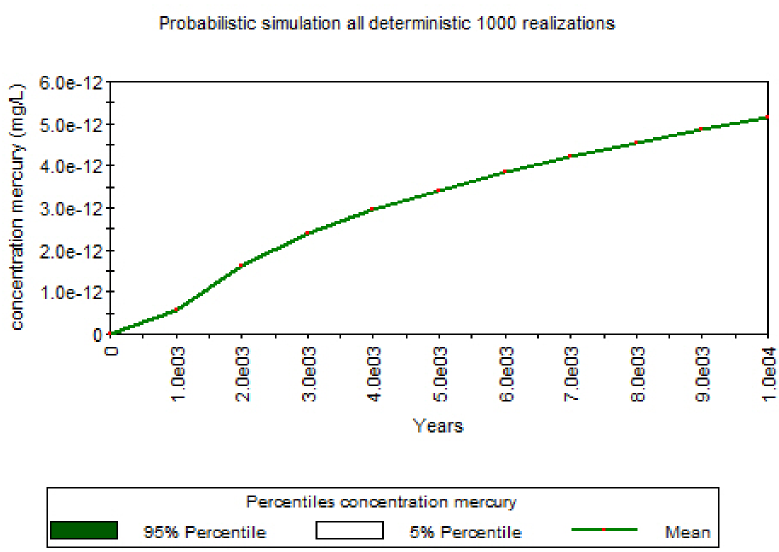

Figure 24 Output concentrations for 1000 realizations

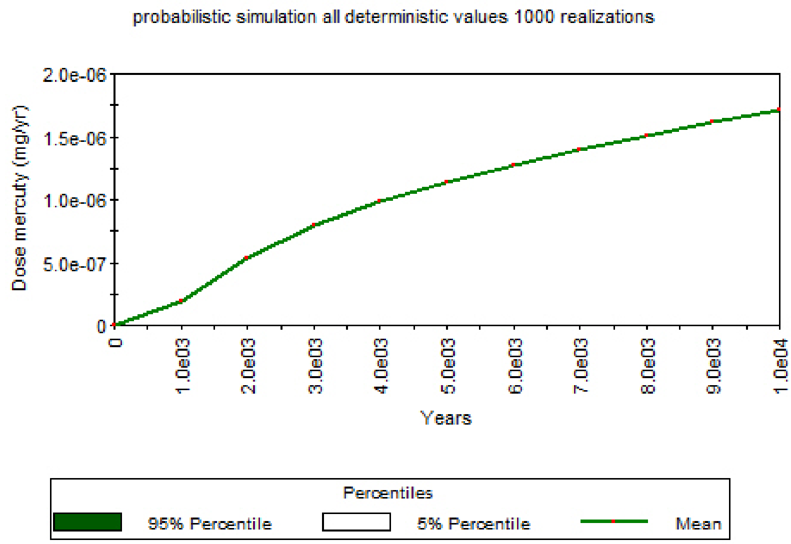

Figure 25 Output dose mercury for 1000 realizations

Figure 25 shows the dose of mercury for 1000 realizations; the 95th percentile was 1.71×10^{-6} mg/yr for 10,000 years.

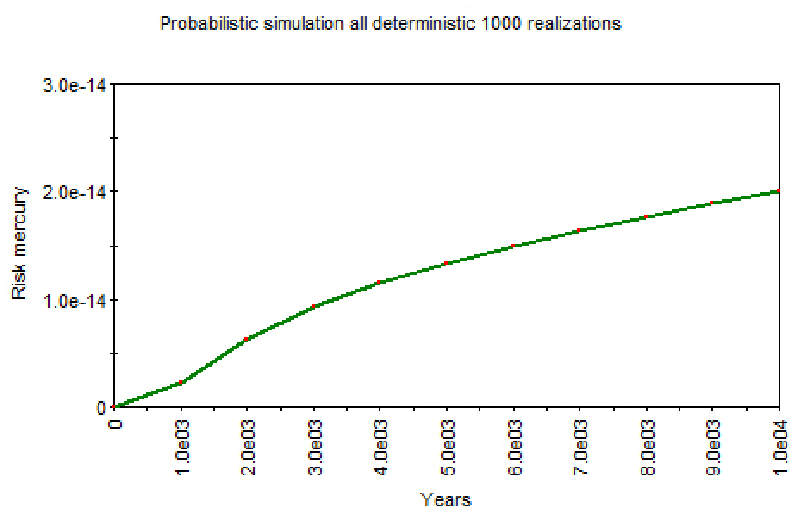

Figure 26 Risk for mercury for 1000 realizations

Figure 26 shows the risk for mercury for 1000 realizations; the percentile 95th was 2.01×10^{-14}.

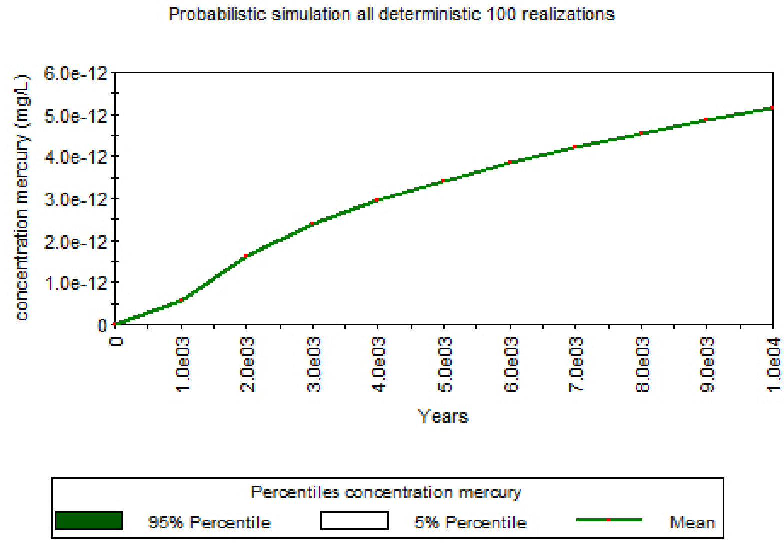

Figure 27 Output concentrations for 100 realizations

Figure 27 shows the concentration of mercury using 100 realizations; the 95th percentile was $5.14x10^{-12}$, the same value for 1000 realizations.

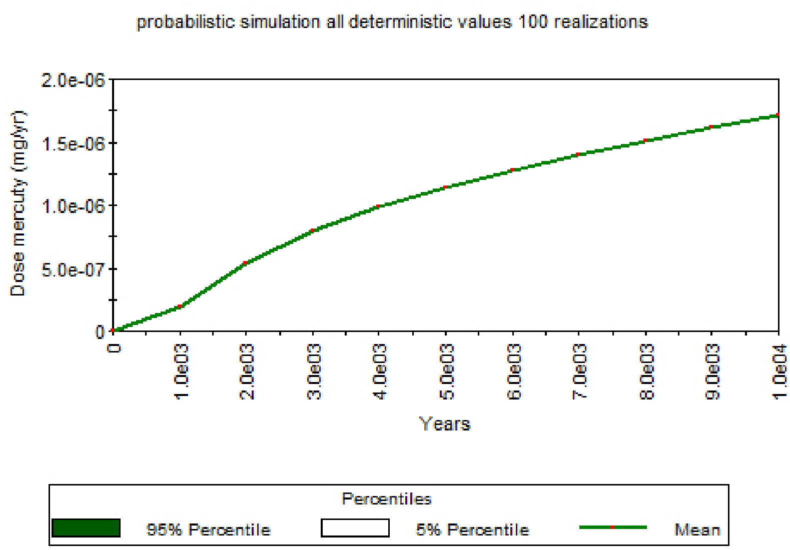

Figure 28 Output dose for 100 realizations

In Figure 28 the dose for mercury concentration in groundwater, using 100 realizations for the simulations, was 1.71×10^{-6} mg/yr for the value of the 95th percentile, which is the same value as used for the 1000 realization simulation.

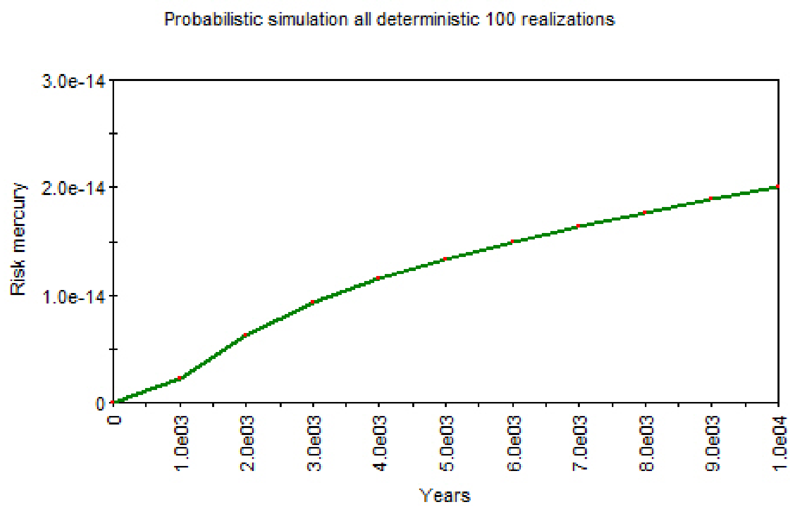

Figure 29 Output risk for 100 realizations

Figure 29 shows the risk for mercury using 100 realizations; the 95th percentile was $2.01 \text{x-} 10^{-14}$, the same as for 1000 realizations.

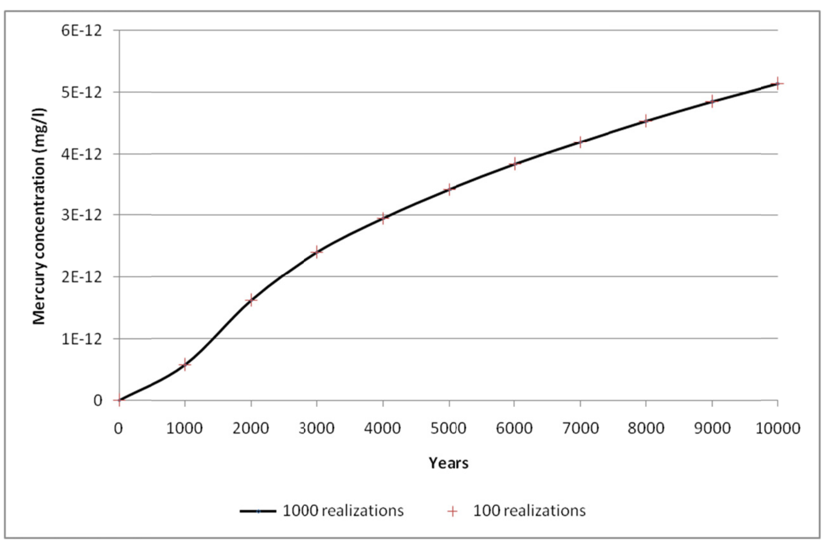

Figure 30 Mercury concentration output for 1000 and 100 realizations

Figure 30 shows both the 95th percentile concentration of mercury for 1000 and 100 realizations. The graph shows there was no difference between the two; therefore, for all probabilistic simulations a set of 100 realizations was selected.

Table 30 reflects the results for the 95th percentile for concentration, dose, and risk for mercury, with a probabilistic simulation using all deterministic input parameters and the setting for 100 realizations.

Table 30 95th percentile for outputs deterministic simulations

Output	**Units**	**95th value**	**Year**
Concentration mercury	mg/L	5.14×10^{-12}	10000
Dose	mg/yr	1.71×10^{-6}	10000
Risk	----	2.01×10^{-14}	10000

Several deterministic simulations were done for a time period of 100 years and for different distances to the drinking water well. The purpose of the variation of the distances from 50 meter until 1000 meters was to evaluate the potential hazard of the contaminated water for a receptor well today. Figure 31 shows the concentration of mercury for distances from the well over a 100 year period. As expected, the concentration of mercury decreases as the distance increases. After 50 meters, a two to three orders of magnitude lower concentration of mercury is expected in the groundwater.

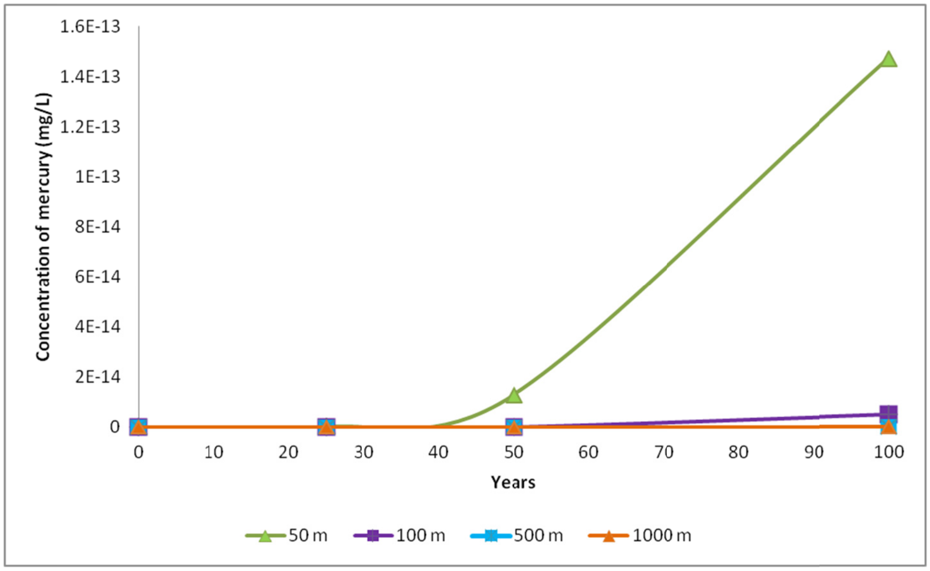

Figure 31 Concentration of mercury for diferent distance to the well (100 years)

Figure 32 shows the concentration of mercury for different distances ranging from 50 to 1000 meters and for 25, 50, and 100 years. For the distance of 50 meters, the concentration increases with time. For distances greater than 50 meters, the concentration has the tendency to decrease with time until the concentration is zero. This shows that the travel distance of the contaminated water to a receptor is an important factor to consider for the potential hazard to humans.

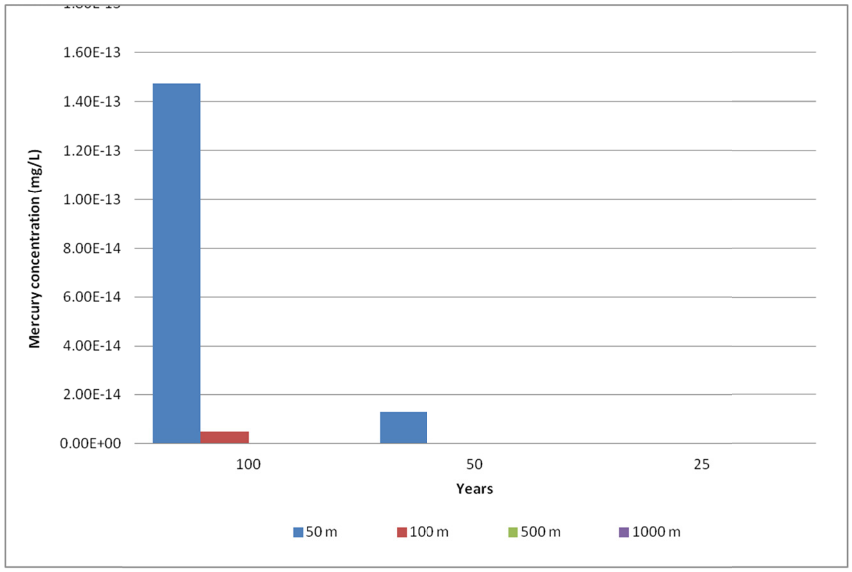

Figure 32 Comparison of concentration for different distance and years

1.16 Risk evaluation infiltration rate probabilistic simulations

To determine the effect of the rate of infiltration, a series of simulations used stochastic input for the infiltration rate parameter and deterministic inputs for the remaining parameters. The infiltration rate was described with a normal distribution with a mean of 0.612 cm/yr and a standard deviation of 0.035 cm/yr. Figure 33 shows the concentration of mercury outputs from GOLDSIM for a probabilistic simulation using a stochastic infiltration rate and 100 realizations.

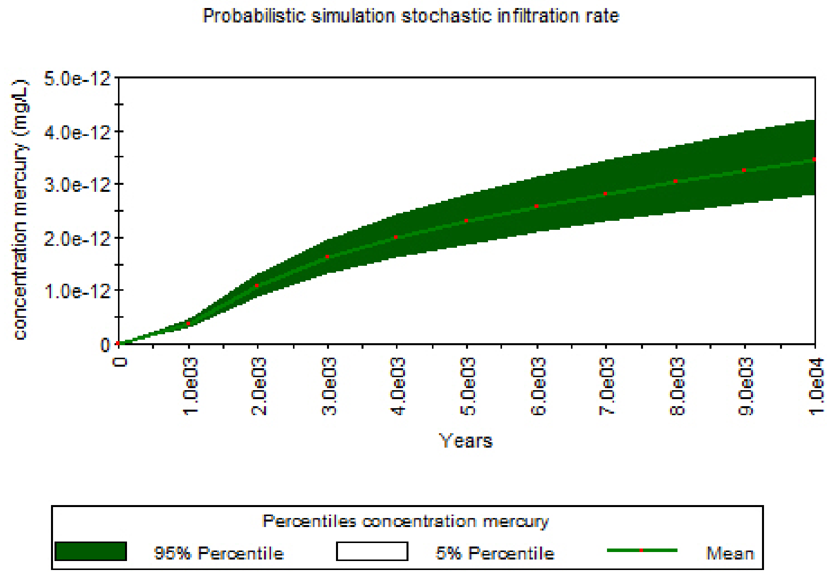

Figure 33 Concentration of mercury with infiltration rate stochastic

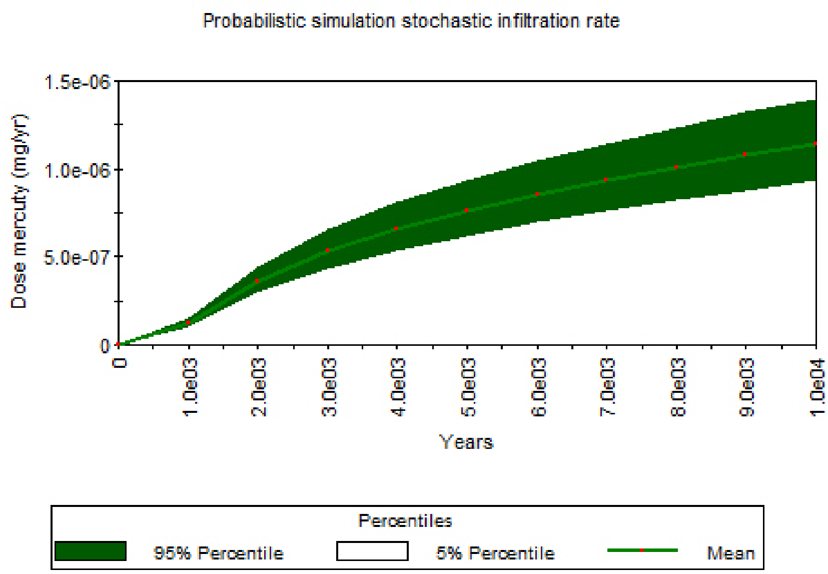

Figure 34 Dose for mercury with infiltration rate stochastic

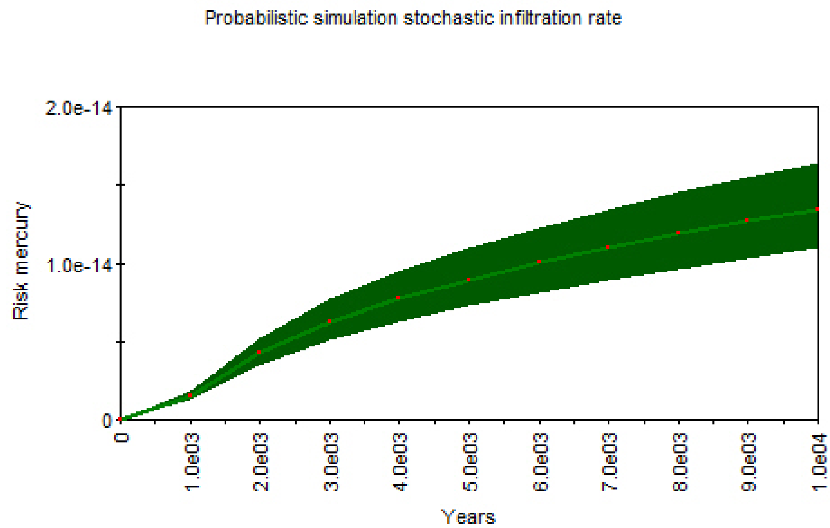

Figure 35 Risk mercury with infiltration rate stochastic

The 95th percentile for concentration, dose, and risk for mercury are shown in Table 31.

Table 31 95th percentile for outputs deterministic simulations

Output	Units	95th value	Year
Concentration mercury	mg/L	4.22×10^{-12}	10000
Dose	mg/yr	1.40×10^{-6}	10000
Risk	----	1.65×10^{-14}	10000

Table 32 shows the percentage of exceedance for concentration, dose, and risk for mercury for infiltration rate stochastic parameter.

Table 32 Output percentage of exceedance for infiltration rate stochastic

Output	Units	Highest	Lowest	Median
Concentration mercury	mg/L	4.46×10^{-12}	2.43×10^{-12}	3.43×10^{-12}
Dose	mg/yr	1.48×10^{-6}	8.07×10^{-7}	1.14×10^{-6}
Risk	----	1.74×10^{-14}	9.48×10^{-15}	1.34×10^{-14}

1.17 Risk evaluation dry bulk density probabilistic simulations

Two sets of simulations were performed using stochastic inputs for the dry bulk density. A normal distribution with mean of 1.067 g/cm3 and variance of 0.017 g/cm^3

were used for the first probabilistic simulation for the source and unsaturated zone. For the second simulation, a Gamma distribution with mean 1.108 g/cm^3 and variance of 0.014 g/cm^3 were used for the saturated zone. For both simulations, the rest of the parameters stayed as deterministic values. Figure 36 shows the outputs from GOLDSIM of the probabilistic simulation for stochastic dry bulk density for unsaturated and source.

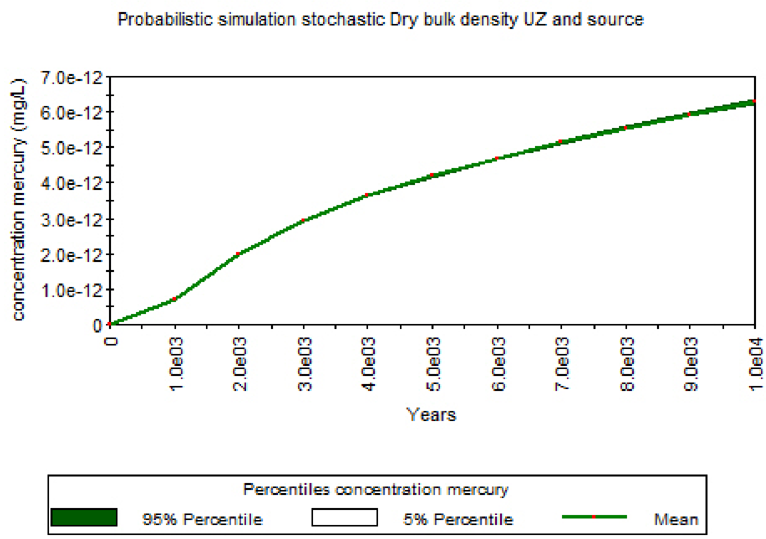

Figure 36 Concentration of mercury for stochastic dry bulk density

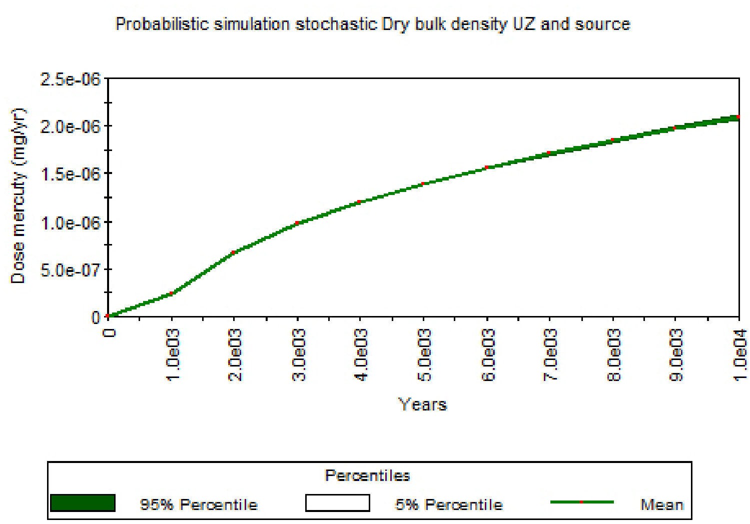

Figure 37 Dose mercury dry bulk density

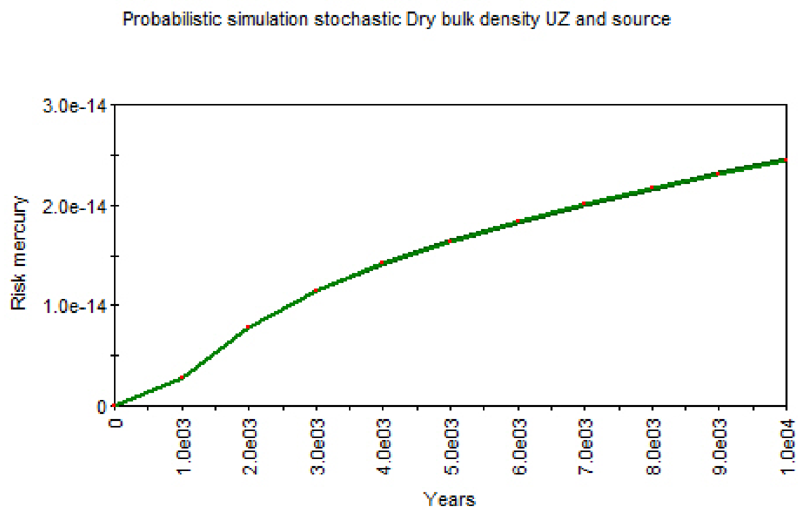

Figure 38 Risk mercury using a stochastic dry bulk density

Table 33 lists the 95th percentile for concentration, dose, and risk for mercury using a stochastic dry bulk density for the source and unsaturated zone.

Table 33 95th with dry bulk density stochastic for unsaturated zone and source

Output	Units	95th value	Year
Concentration mercury	mg/L	6.37×10^{-12}	10000
Dose	mg/yr	2.12×10^{-6}	10000
Risk	----	2.49×10^{-14}	10000

The percentage of exceedance for mercury concentration, dose, and risk for infiltration rate stochastic parameter is shown in Table 34.

Table 34 Percentage of exceedance for dry bulk density stochastic for UZ

Output	Units	Highest	Lowest	Median
Concentration	mg/L	6.42×10^{-12}	6.16×10^{-12}	6.29×10^{-12}
Dose	mg/yr	2.14×10^{-6}	2.05×10^{-6}	2.09×10^{-6}
Risk	----	2.51×10^{-14}	2.40×10^{-14}	2.45×10^{-14}

Figure 39 shows the concentration of mercury as an output in GOLDSIM for the probabilistic simulation using a stochastic dry bulk density for the saturated zone. A gamma distribution with a mean of 1.108 g/cm^3 and variance of 0.014 g/cm^3 was used.

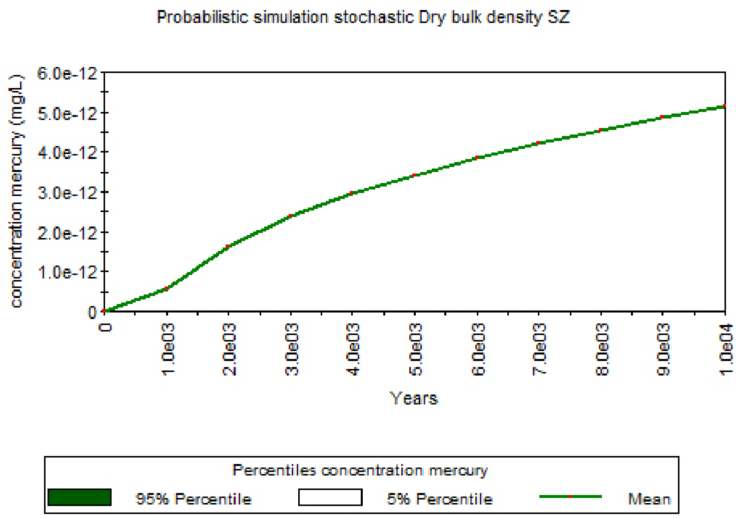

Figure 39 Concentration of mercury using a stochastic dry bulk density SZ

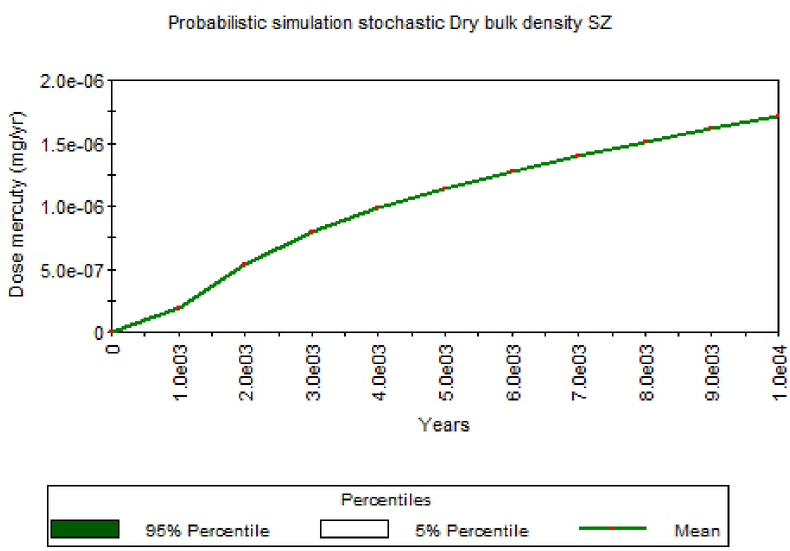

Figure 40 Dose mercury using a stochastic dry bulk density SZ

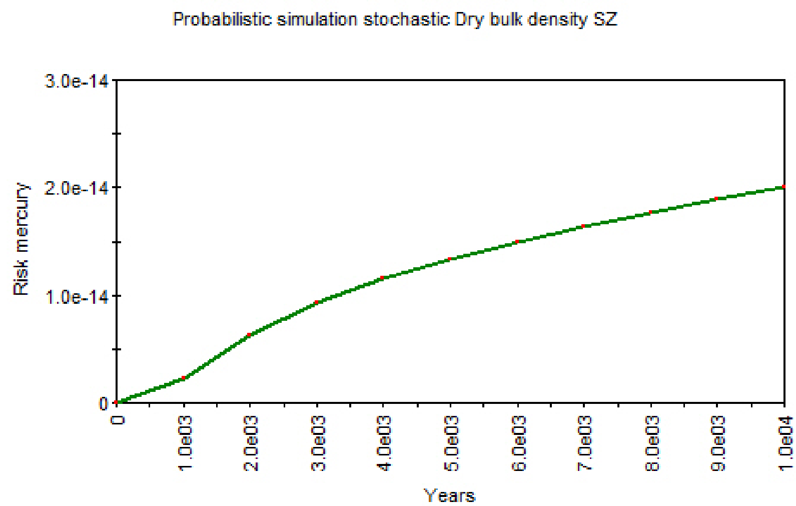

Figure 41 Risk mercury using a stochastic dry bulk density SZ

Table 35 shows the 95th percentile for concentration, risk, and dose of mercury, using a stochastic dry bulk density for the saturated zone.

Table 35 95th percentile for outputs with dry bulk density stochastic for SZ

Output	Units	95th value	Year
Concentration mercury	mg/L	$5.14x10^{-12}$	10000
Dose	mg/yr	$1.71x10^{-6}$	10000
Risk	----	$2.01x10^{-14}$	10000

The percentage of exceedance for mercury concentration, dose, and risk for a stochastic dry bulk density for the saturated zone is show in Table 36.

Table 36 Output percentage of exceedance for dry bulk density stochastic for SZ.

Output	Units	Highest	Lowest	Median
Concentration	mg/L	$5.142x10^{-12}$	$5.142x10^{-12}$	$5.142x10^{-12}$
Dose	mg/yr	$1.71x10^{-6}$	$1.71x10^{-6}$	$1.71x10^{-6}$
Risk	----	$2.01x10^{-14}$	$2.01x10^{-14}$	$2.01x10^{-14}$

1.18 Risk evaluation porosity probabilistic simulations

Two sets of simulations were performed for the stochastic porosity. The first simulation was for the source and unsaturated zone using a Gamma distribution for porosity with a mean of 0.487 and variance of 0.1333, the rest of the input parameters stayed deterministic. Figure 42 shows the outputs form GOLDSIM of the probabilistic simulation for stochastic dry bulk density for unsaturated and source.

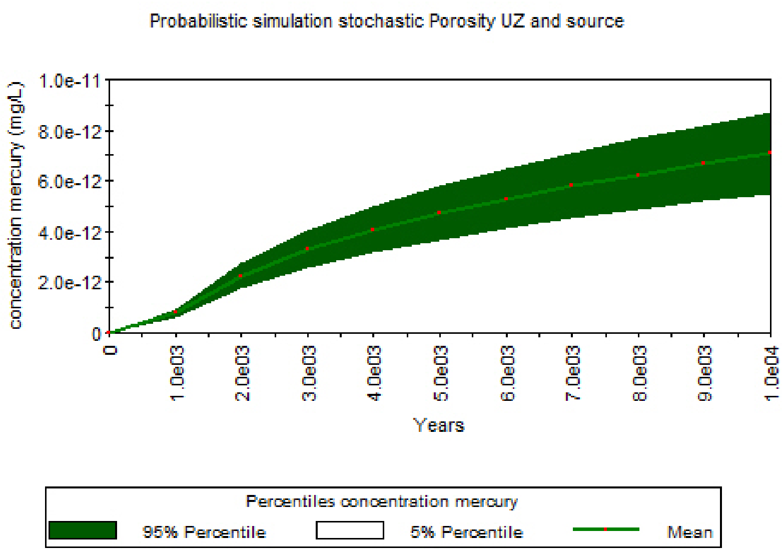

Figure 42 Concentration of mercury using a stochastic porosity source and UZ

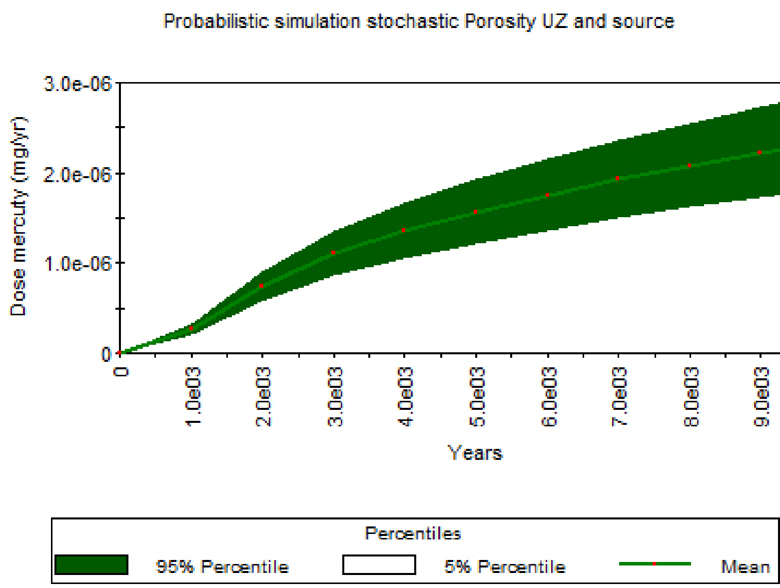

Figure 43 Dose for mercury using a stochastic porosity source and UZ

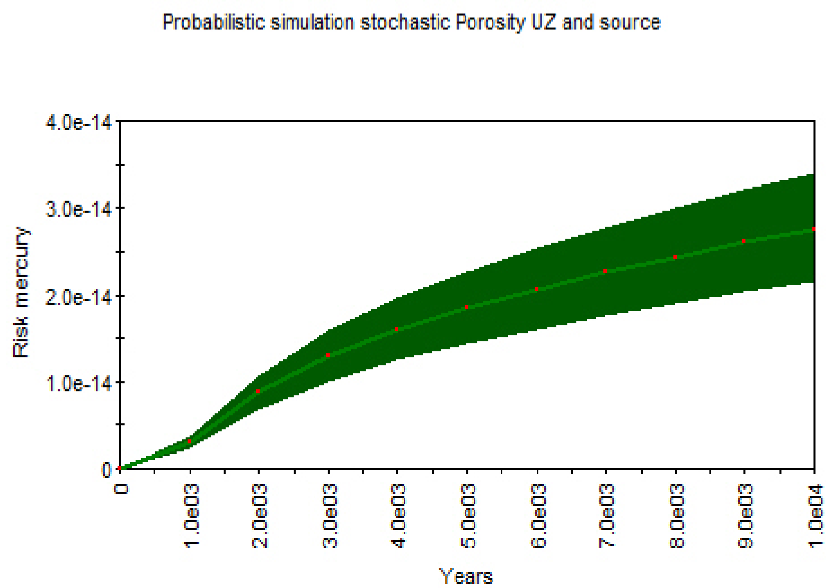

Figure 44 Risk for mercury using a stochastic porosity source and UZ

Table 37 shows the 95th percentile for concentration, dose, and risk of mercury, using a stochastic porosity for the source and unsaturated zone.

Table 37 95th percentile for outputs with porosity stochastic for UZ and source

Output	Units	95th value	Year
Concentration mercury	mg/L	8.70×10^{-12}	10000
Dose	mg/yr	2.89×10^{-6}	10000
Risk	----	3.40×10^{-14}	10000

The percentage of exceedance for mercury concentration, dose, and risk for infiltration rate stochastic parameter is shown in Table 38.

Table 38 Output percentage of exceedance for porosity stochastic for UZ

Output	Units	Highest	Lowest	Median
Concentration	mg/L	9.54×10^{-12}	4.92×10^{-12}	7.08×10^{-12}
Dose	mg/yr	3.17×10^{-6}	1.64×10^{-6}	2.35×10^{-6}
Risk	----	3.72×10^{-14}	1.92×10^{-14}	2.67×10^{-14}

For the second simulation, a stochastic porosity for the saturated zone was used with a lognormal distribution with a mean of 0.134 and variance of 0.014. Figure 45 shows the concentration of mercury as outputs from GOLDSIM.

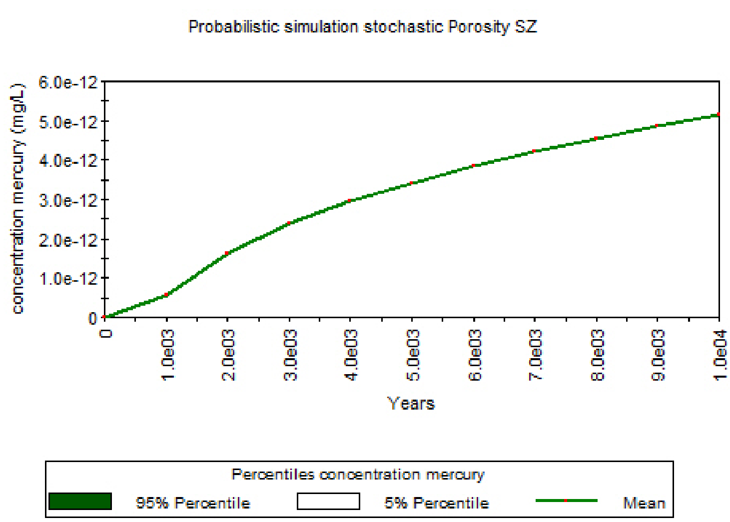

Figure 45 Concentration of mercury using a stochastic porosity SZ

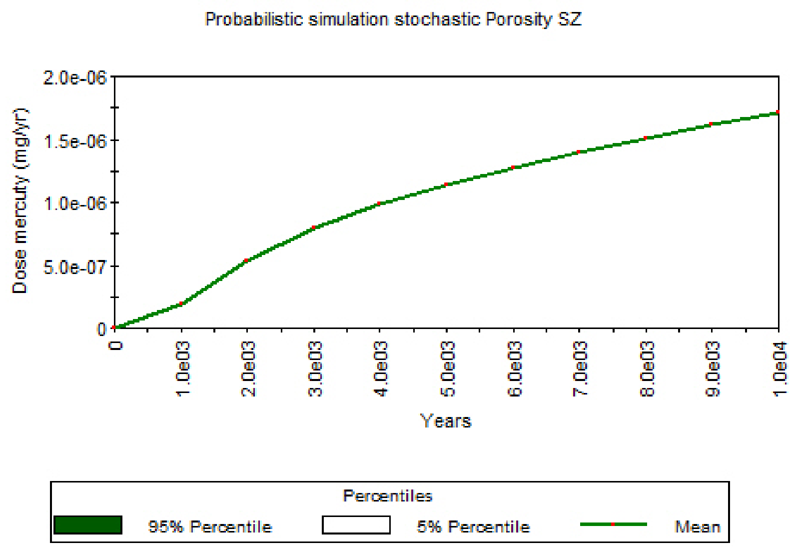

Figure 46 Dose for mercury using a stochastic porosity for SZ

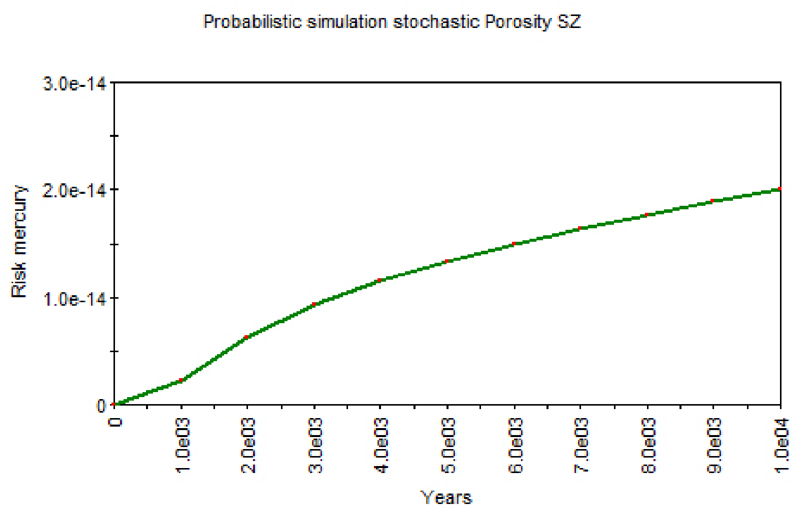

Figure 47 Risk for mercury using a stochastic porosity for SZ

Table 39 shows the 95th percentile for concentration, dose, and risk of mercury, for a probabilistic simulation using a stochastic porosity in the saturated zone.

Table 39 95th percentile for outputs with porosity stochastic for SZ

Output	Units	95th value	Year
Concentration mercury	mg/L	$5.14x10^{-12}$	10000
Dose	mg/yr	$1.71x10^{-6}$	10000
Risk	----	$2.01x10^{-14}$	10000

The percentage of exceedance for mercury concentration, dose, and risk for infiltration rate stochastic parameter is shown in Table 40.

Table 40 Output percentage of exceedance for porosity stochastic for UZ

Output	Units	Highest	Lowest	Median
Concentration	mg/L	$5.14x10^{-12}$	---	---
Dose	mg/yr	$1.71x10^{-6}$	----	----
Risk	----	$2.01x10^{-14}$	---	----

1.19 Risk evaluation partition coefficient probabilistic simulations

Three sets of simulations were performed using a stochastic input for the partition coefficient. The first simulation was for the mercury containment system using a normal distribution with a mean of 468.75 ml/g and a variance of 4.35 (log of the variance 22747.3). The second simulation was for the unsaturated zone using a lognormal distribution with a mean of 65 and a variance of 2.95 (log of the variance 907.184). The third simulation was for the saturated zone using a lognormal distribution with a mean of 133.32 and a variance of 3.11 (log of variance 1294.12), the rest of the input parameters stayed deterministic.

Figure 48 shows the concentration of mercury as an output from GOLDSIM of the probabilistic simulation for stochastic partition coefficient for the source, using a normal distribution.

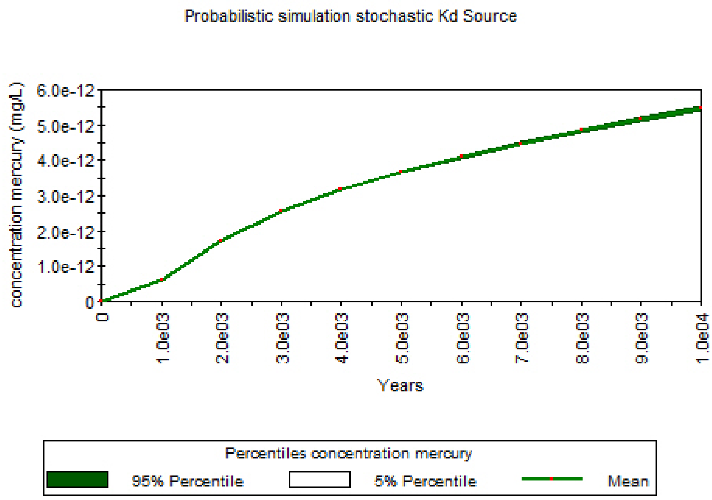

Figure 48 Concentration of mercury using a stochastic Kd for the source

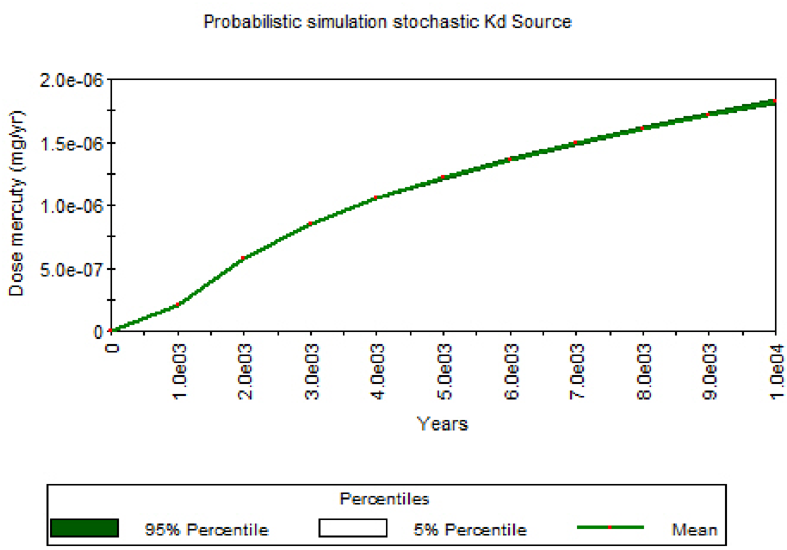

Figure 49 Dose of mercury using a stochastic Kd for the source

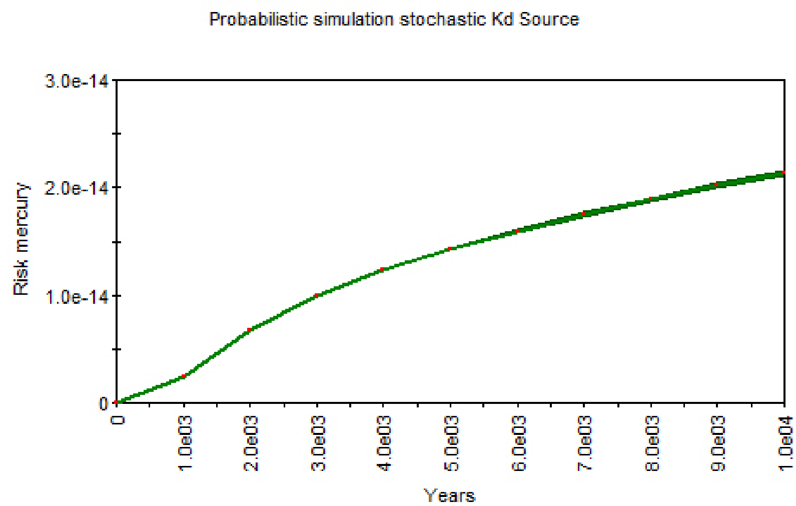

Figure 50 Risk of mercury using a stochastic Kd for the source

Table 41 shows the 95th percentile for concentration, dose, and risk for mercury, using a stochastic partition coefficient for the source.

Table 41 95th percentile for partition coefficient stochastic for source

Output	Units	95th value	Year
Concentration mercury	mg/L	5.56×10^{-12}	10000
Dose	mg/yr	1.85×10^{-6}	10000
Risk	----	2.17×10^{-14}	10000

The percentage of exceedance for mercury concentration, dose, and risk using a stochastic partition coefficient are shown in Table 42.

Table 42 Output percentage of exceedance for partition coefficient for source

Output	Units	Highest	Lowest	Median
Concentration	mg/L	5.60×10^{-12}	5.32×10^{-12}	5.48×10^{-12}
Dose	mg/yr	1.86×10^{-6}	1.77×10^{-6}	1.82×10^{-6}
Risk	----	2.18×10^{-14}	2.08×10^{-14}	2.14×10^{-14}

For the second probabilistic simulation in GOLDSIM a stochastic partition coefficient with a lognormal distribution with a mean of 65 and a variance of 2.95 (log of the variance 907.184) was used. Figure 51 shows the concentration of mercury, an output from GOLDSIM of the probabilistic simulation for stochastic partition coefficient for the unsaturated zone, using a lognormal distribution.

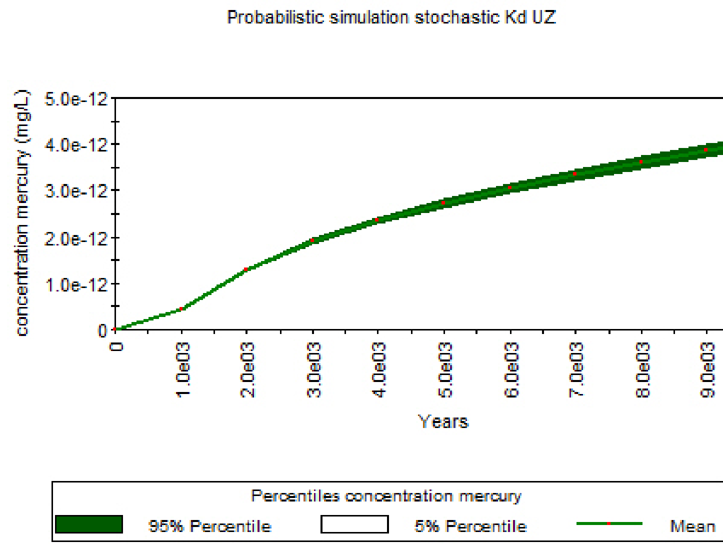

Figure 51 Concentration of mercury using a stochastic Kd for the UZ

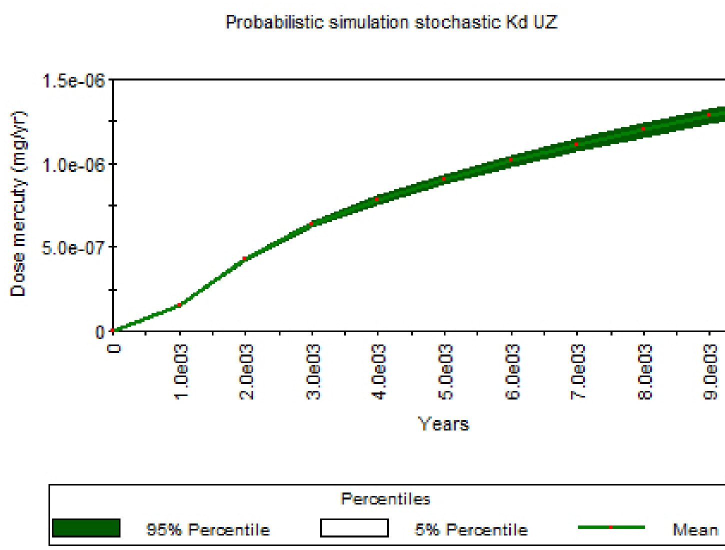

Figure 52 Dose for mercury using a stochastic Kd for UZ

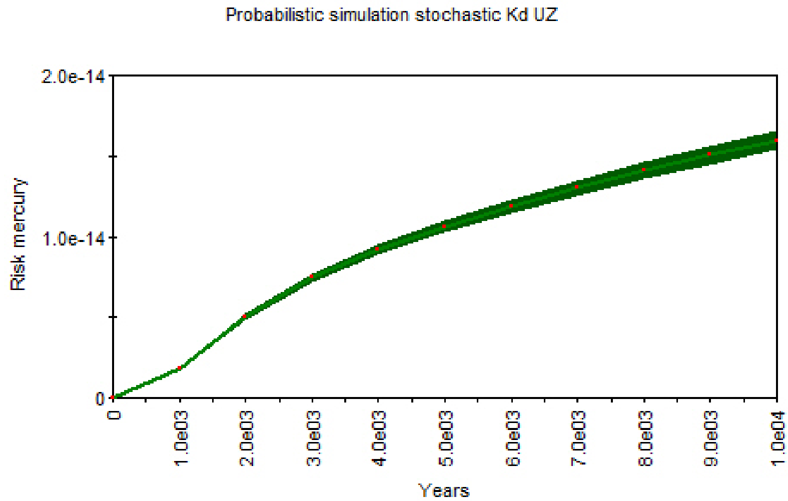

Figure 53 Risk for mercury using a stochastic Kd for UZ

Table 43 shows the 95th percentile for concentration, dose, and risk for mercury, using a stochastic partition coefficient for the unsaturated zone.

Table 43 95th percentile for outputs with partition coefficient stochastic for UZ

Output	Units	95th value	Year
Concentration mercury	mg/L	4.24×10^{-12}	10000
Dose	mg/yr	1.41×10^{-6}	10000
Risk	----	1.65×10^{-14}	10000

The percentage of exceedance for mercury concentration, dose, and risk using a stochastic partition coefficient are shown in Table 44.

Table 44 Output percentage of exceedance for partition coefficient for UZ

Output	Units	Highest	Lowest	Median
Concentration	mg/L	4.34×10^{-12}	3.87×10^{-12}	4.09×10^{-12}
Dose	mg/yr	1.44×10^{-6}	1.28×10^{-6}	1.36×10^{-6}
Risk	----	1.69×10^{-14}	1.51×10^{-14}	1.59×10^{-14}

For the third probabilistic simulation in GOLDSIM a stochastic partition coefficient with a lognormal distribution with a mean of 133.32 and a variance of 3.11

(log of variance 1294.12) was used. Figure 54 shows the concentration of mercury, an outputs from GOLDSIM of the probabilistic simulation for stochastic partition coefficient for the saturated zone using a lognormal distribution.

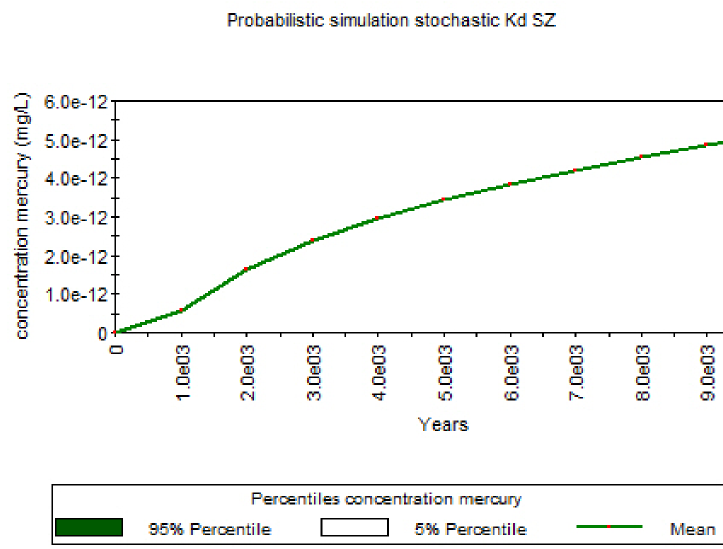

Figure 54 Concentration of mercury using a stochastic Kd for the SZ

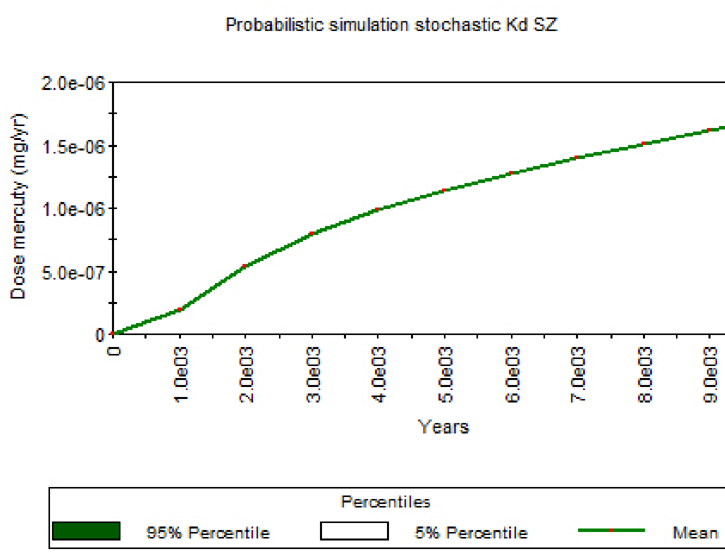

Figure 55 Dose of mercury using a stochastic Kd for the SZ

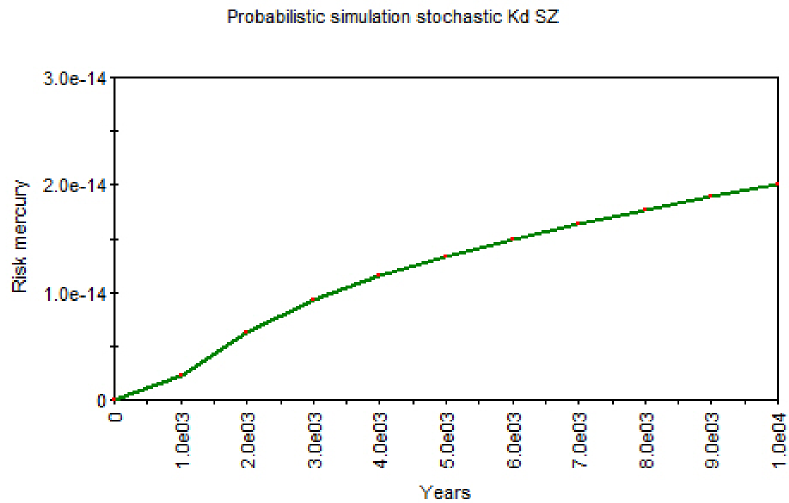

Figure 56 Risk of mercury using a stochastic Kd for the SZ

Table 45 shows the 95th percentile for concentration, dose, and risk for mercury, using a stochastic partition coefficient for the saturated zone.

Table 45 95th percentile for outputs with partition coefficient stochastic for SZ

Output	Units	95th value	Year
Concentration mercury	mg/L	5.14×10^{-12}	10000
Dose	mg/yr	1.71×10^{-6}	10000
Risk	----	2.01×10^{-14}	10000

The percentage of exceedance for mercury concentration, dose, and risk using a stochastic partition coefficient are shown in Table 46.

Table 46 Output percentage of exceedance for partition coefficient for SZ

Output	Units	Highest	Lowest	Median
Concentration	mg/L	5.14×10^{-12}	----	----
Dose	mg/yr	1.71×10^{-6}	----	----
Risk	----	2.01×10^{-14}	----	----

Table 47 shows the percentage of uncertainty variance for all the simulations. The highest uncertainty variance for the concentration of mercury occurred when the stochastic porosity in the unsaturated zone and source was used with a value of 58%.

Second is the infiltration rate with an uncertainty of 51%. For the stochastic parameters dry bulk density unsaturated zone and source, porosity in the saturated zone and partition coefficient in the saturated zone did not have variance, which means these parameters do not have an influence on the concentration of mercury.

Table 47 Percentage of uncertainty variance simulations

Simulation for concentration of mercury	Percentage of uncertainty variance
All inputs deterministic	0
Stochastic infiltration rate	51
Stochastic dry bulk density unsaturated zone and source	3
Stochastic dry bulk density saturated zone	0
Stochastic porosity unsaturated zone and source	58
Stochastic porosity saturated zone	0
Partition coefficient (Kd) source	3
Partition coefficient (Kd) unsaturated zone	7
Partition coefficient (Kd) saturated zone	0

1.20 Comparison of simulations

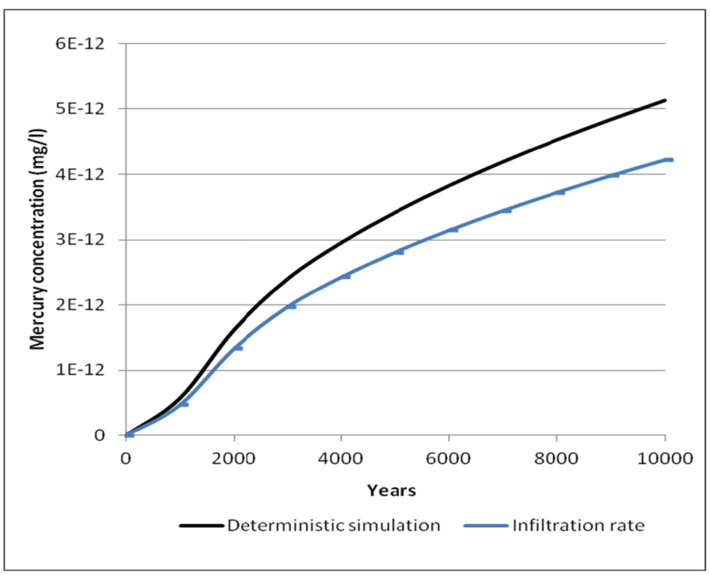

Figure 57 Simulations of deterministic and stochastic infiltration rate

Figure 57 compares the 95th percentile of the concentration of mercury of the simulations deterministic and stochastic infiltration rate. The graph illustrates that the values for stochastic infiltration rate are below the values for the deterministic simulation.

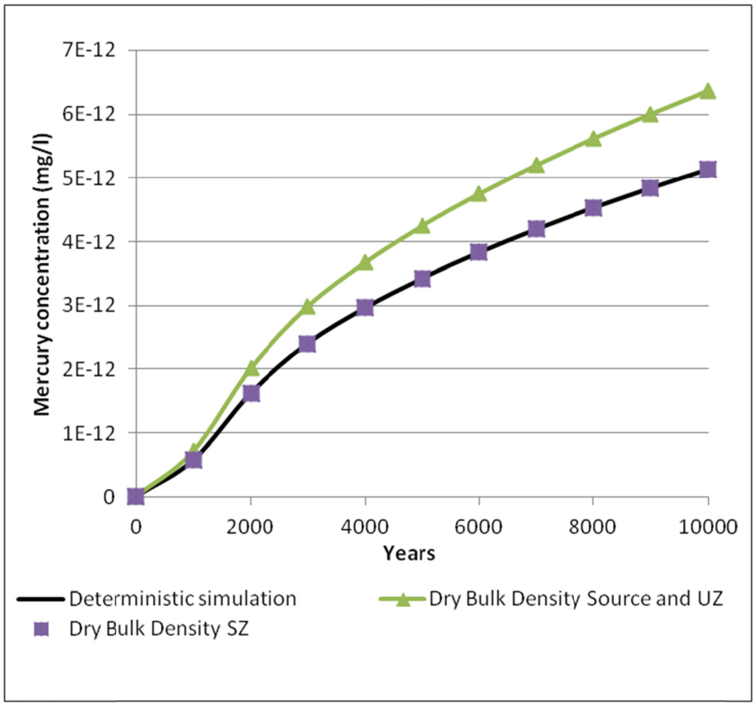

Figure 58 Simulations deterministic and stochastic dry bulk UZ and SZ

Figure 58 compares the 95th percentile of the concentration of mercury of the simulations deterministic and stochastic dry bulk density unsaturated and saturated zones. The graph illustrates that the values for stochastic dry bulk density unsaturated zone are higher than the values for the deterministic simulation, and dry bulk density saturated zone is the same as deterministic.

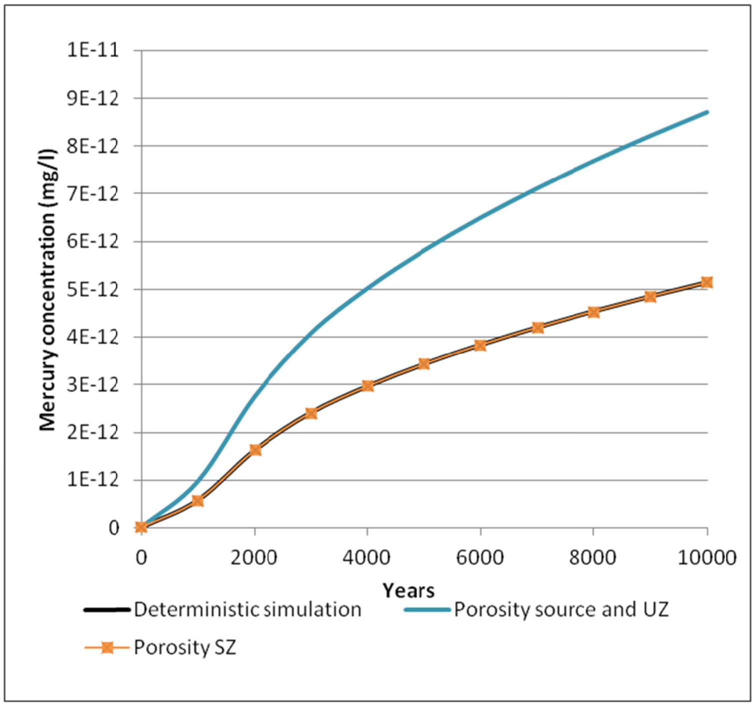

Figure 59 Simulations deterministic and stochastic porosity UZ and SZ

Figure 59 compares the 95th percentile of the concentration of mercury of the simulations deterministic and stochastic porosity unsaturated and saturated zones. The graph illustrates that the values for stochastic porosity unsaturated zone are higher than the values for the deterministic simulation, and dry bulk density saturated zone is the same as deterministic.

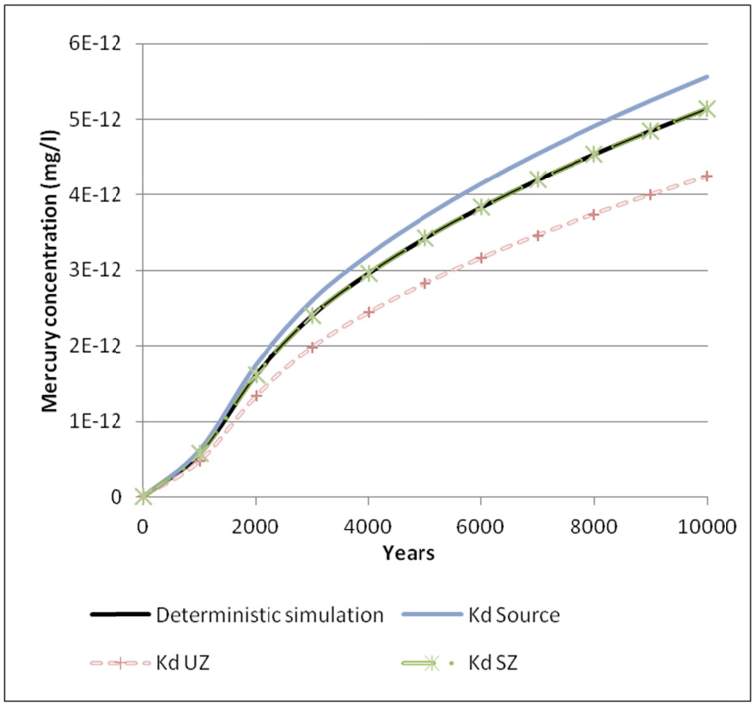

Figure 60 Simulations deterministic and stochastic Kd source, UZ and SZ

Figure 60 compares the 95th percentile of the concentration of mercury of the simulations deterministic and the probabilistic simulation. The probabilistic simulation used a stochastic partition coefficient source, unsaturated and saturated zone. The graph illustrates that the values for stochastic partition coefficient source are higher than the values for the deterministic simulation, that the partition coefficient saturated zone is the same as deterministic, and that the partition coefficient unsaturated zone is below the values for deterministic simulation.

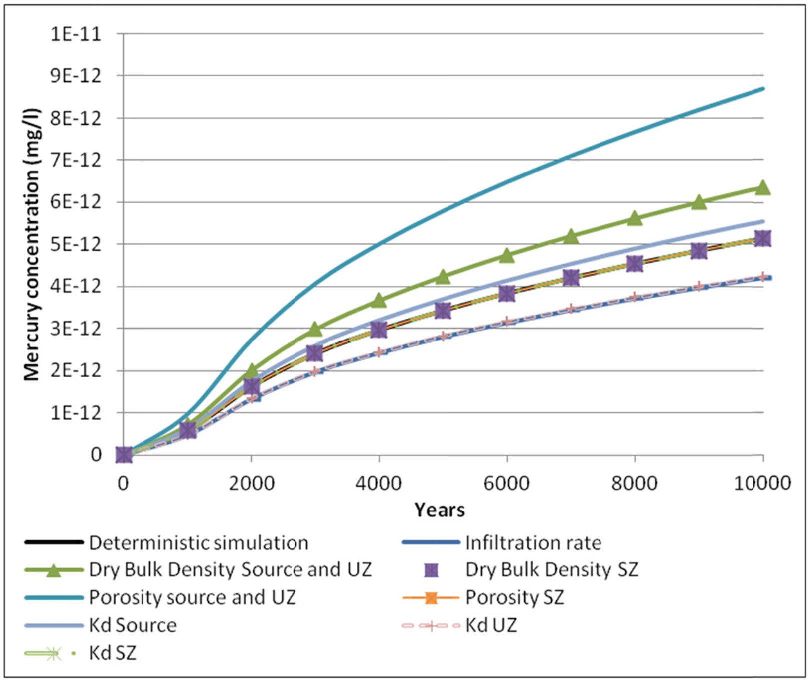

Figure 61 Simulations for all selected parameters

Figure 61 shows the 95th percentile of the concentration of mercury for all of the simulations.

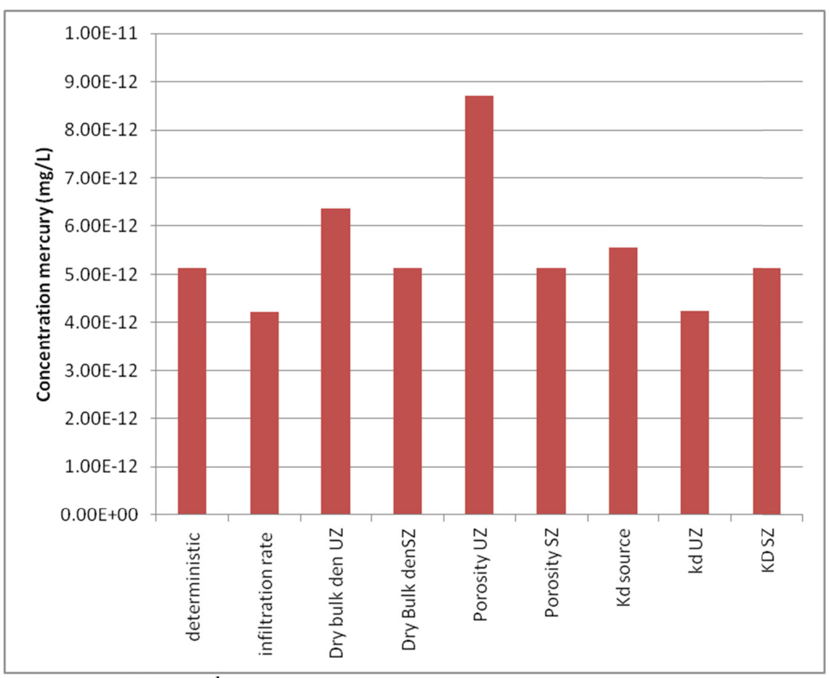

Figure 62 Comparison 95th percentile concentration of mercury for simulations

Figure 62 shows the 95th percentile values for all of the simulations for concentration of mercury. The simulations with all the value set as deterministic or single value have the deterministic simulation with a value of 5.14×10^{-12} mg/L, which was used as a reference point to compare with all the simulations of the stochastic simulations. The highest value for concentration of mercury in comparison with the reference value was the simulation for stochastic porosity in the source and unsaturated zone with a value of 8.70×10^{-12}, which has a ratio of 1.69. The lowest value for the concentration of mercury was found in two simulations, one for stochastic infiltration rate and the other for the stochastic partition coefficient for the unsaturated zone, both with a ratio of 0.82.

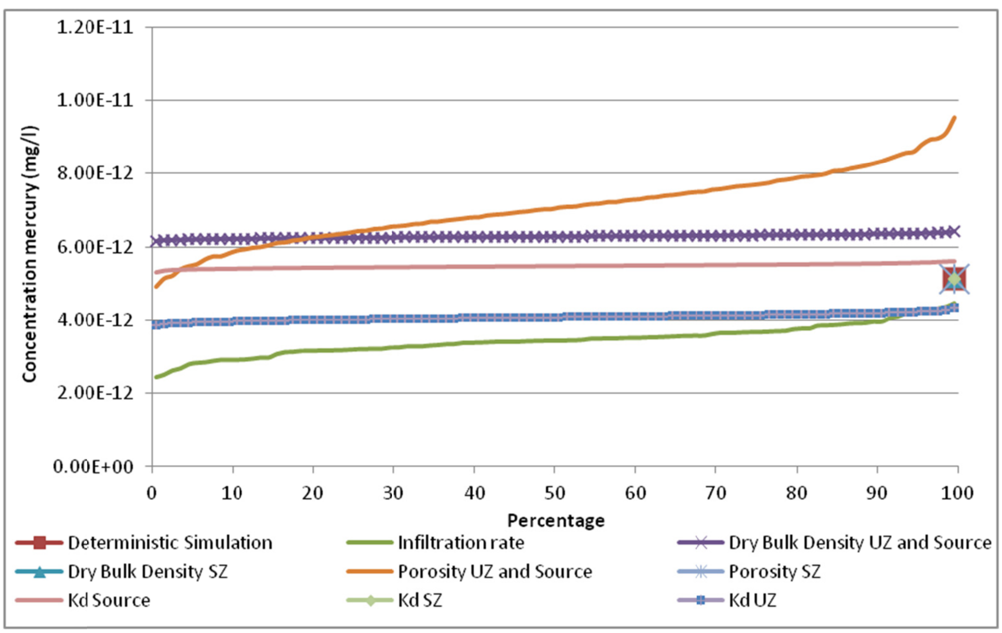

Figure 63 Percentage of exceedance for all simulations

Figure 63 shows the percentage of exceedance for all stochastic simulations compared with the deterministic value for concentration of mercury.

Table 48 lists the result for the 95th percentile for the concentration of mercury, dose, and risk for each of the simulations for 10,000 years, applying the different distributions for the selected stochastic parameters.

Table 48 Summary of the simulations for 10,000 years

Type of Simulation	**Type of distribution**	**Concentration 95th (mg/L)**	**Dose 95th (mg/yr)**	**Risk 95th**
Deterministic	-----	5.14×10^{-12}	1.71×10^{-6}	2.01×10^{-14}
Infiltration rate	normal	4.22×10^{-12}	1.40×10^{-6}	1.65×10^{-14}
Dry bulk density (source and UZ)	normal	6.37×10^{-12}	2.12×10^{-6}	2.49×10^{-14}
Dry bulk density (SZ)	Gamma	5.14×10^{-12}	1.71×10^{-6}	2.01×10^{-14}
Porosity (source and SZ)	Gamma	8.70×10^{-12}	2.89×10^{-12}	3.40×10^{-14}
Porosity (SZ)	lognormal	5.14×10^{-12}	1.71×10^{-6}	2.01×10^{-14}
Kd source	normal	5.56×10^{-12}	1.85×10^{-6}	2.17×10^{-14}
Kd UZ	lognormal	4.24×10^{-12}	1.41×10^{-6}	1.69×10^{-14}
Kd SZ	lognormal	5.14×10^{-12}	1.71×10^{-6}	2.01×10^{-14}

Table 49 Summary of distributions used for the simulations

Type of Simulation	**Type of distribution**	**Mean (µ)**	**Standard deviation**
Deterministic	-----	----	-----
Infiltration rate	normal	0.61	0.036
Dry bulk density (source and UZ)	normal	1.067	0.017
Dry bulk density (SZ)	Gamma	1.0675	0.019
Porosity (source and SZ)	Gamma	0.487	0.133
Porosity (SZ)	lognormal	0.134	0.014
Kd source	normal	468	22747
Kd UZ	lognormal	65	907
Kd SZ	lognormal	133	1294.12

1.21 Sensitivity analysis of the probabilistic parameters

The contaminant transport of mercury from the source to the selected well GW-904 is a complex process due to many hydrological and transport variables involved with governing the transport and the high degree of inherent uncertainty for each of them. These variables have an impact on the quantity of mercury present in the well, and furthermore on the risk and dose which can affect the health of the receptor. A sensitivity analysis was conducted for each of the stochastic parameters to assess the influence or relative importance of these input variables to the output, which is the concentration of mercury in the well.

GOLDSIM provides statistical sensitivity analyses through the multivariate result element. The measures that GOLDSIM computes are the coefficient of determination, correlation coefficient, standardized regression coefficient (SRC) partial correlation coefficient, and importance measure. For a risk assessment model many other input variables will have an impact on the overall uncertainty of mercury transport, however this study focused on selected hydrological and transport parameters.

Table 50 shows the results of the sensitivity analysis for all the simulations with stochastic parameters. Measurement of importance varies between 0 and 1, and represents the fraction of the results variance that is explained by the variable. This measure is useful in identifying nonlinear, non-monotonic relationships between an input variable and the result. The concentration of mercury in the drinking water well is equally sensitive to stochastic porosity in the unsaturated and saturated zones, with almost the same measure of importance, 0.81 and 0.80 respectively. The variables with the lowest measures of importance were the infiltration rate, dry bulk density source, and porosity source and partition coefficient in the saturated zone. The coefficient of determination varies between 0 and 1, and represents the fraction of the total variance in the result that can be explained with a linear relationship to the input variables. The closer the value is to 1, the more significant is the relationship between the result and the variables. The dry bulk density parameter for source and UZ, porosity source and UZ, and partition coefficient (Kd) for source and UZ have strong correlations with the concentration of mercury. The correlation coefficient ranges between -1 and 1. A value of 1 implies that a linear equation describes the correlation between the results and the variables; when the results increase the variables increase. A value of -1 implies that all data points lie on a line from which results decrease as variables increase. A value of 0 implies that there is no linear correlation between the result and the variables. The dry bulk density for UZ, the partition coefficient (Kd) source, and the Kd for UZ have a correlation value of -1. The porosity in the UZ has a value of 1, therefore it has a direct correlation with the concentration of mercury output. The Standarized Regression Coefficient (SRC) range between -1 and 1 and provide a normalized measure of the linear relationship between variables and the result. Partial Correlation Coefficient vary between -1 and 1, and reflect the extent to which there is a linear relationship between the selected result and an input variable, after removing the effects of any linear relationships between the other input variables and both the result and the input variable in question.

Table 50 Sensitivity analysis for stochastic parameters

Stochastic parameter	Coefficient of determination	Corr. Coeff.	SRC	Partial coeff.	Importance
Infiltration rate	0.30	0.54	0.54	0.54	0.25
Dry bulk density source	1	0.09	0.001	0.052	0.06
Dry bulk density UZ	1	-1.000	-1.000	-1.000	0.78
Dry bulk density SZ	0	0.000	-0.000	-0.000	unavailable
Porosity source	0.99	0.11	-0.005	-0.051	0.05
Porosity UZ	0.99	1	1	1	0.81
Porosity SZ	0	0.000	-0.000	-0.000	unavailable
Kd source	1	-1.000	-1.000	-1.000	0.77
Kd UZ	1	-1	-1	-1	0.80
Kd SZ	0	-0.000	-0.000	-0.000	0.0000

1.22 Discussion

The deterministic simulation was calculated first to be used as a reference to determine how the concentration of mercury in the well will vary when making simulations for different normal distributions of the selected parameters: infiltration rate, dry bulk density, porosity and partition coefficient.

Based on the results of the previous simulations deterministic and probabilistic and the sensitivity analysis, further simulations were necessary. These new simulations were performed to evaluate how changes of order of magnitude of specific values will have an impact on the concentration of mercury for 10,000 years. These parameters are the initial concentration of mercury in the containment system, the Q of water, which depends on the value of hydraulic conductivity, the log of partition coefficient for the source, unsaturated and saturated zone, and variations of solubility.

A probabilistic simulations was performed for the stochastic porosity in the source and unsaturated zone to evaluate how sensitive this stochastic parameter becomes with the changes of order of magnitude of these selected parameters. The first deterministic simulation included the calculation of concentration of mercury, dose, and risk for a time period of 100 years and different distances to the drinking well. The initial concentration of mercury in the containment system, which was 1 mg/kg (1 ppm), was increased to a value of 60 mg/kg (60 ppm). Figure 64 shows the values of 95th percentiles for concentration of mercury for an initial concentration of mercury of 60 ppm for different distances and a time period of 100 years.

The output concentrations for the different distances for a concentration of 60 ppm are higher compared with the first simulation with a initial concentration of 1 ppm. Both concentrations give an output concentration for mercury in the drinking water well below the MCL, which is 0.002 mg/L for mercury.

Figure 64 shows the 95th percentiles for the concentration of mercury, for different distance to the drinking well and time of 100 years. For the distance of 50 meter the concentration value were 8.84×10^{-12} mg/L, for 100 meter, was 3.03×10^{-13} mg/L, for 500 meter was 1.83×10^{-25} mg/L and for 1000 meter was 0 mg/L.

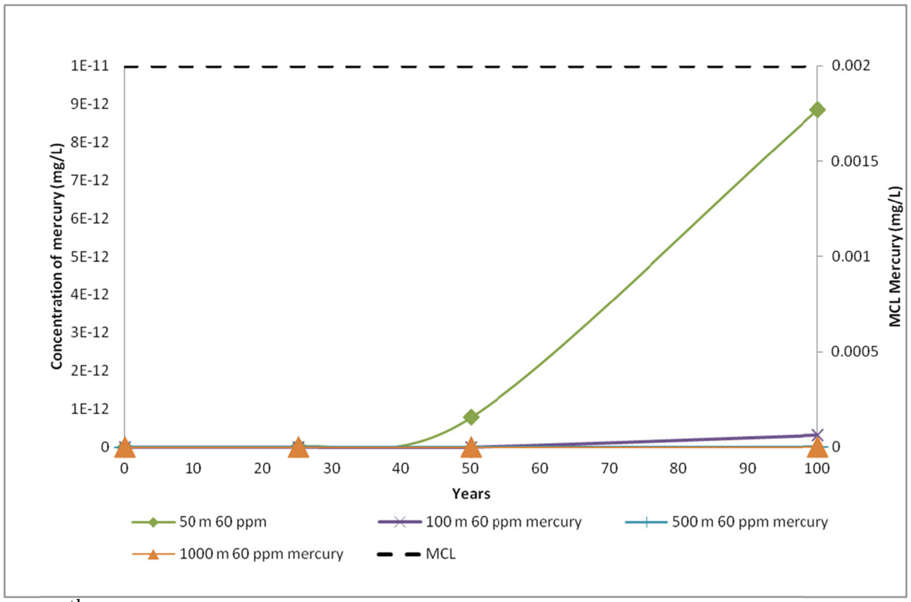

Figure 64 95th concentration of mercury for different distance to well (60 ppm)

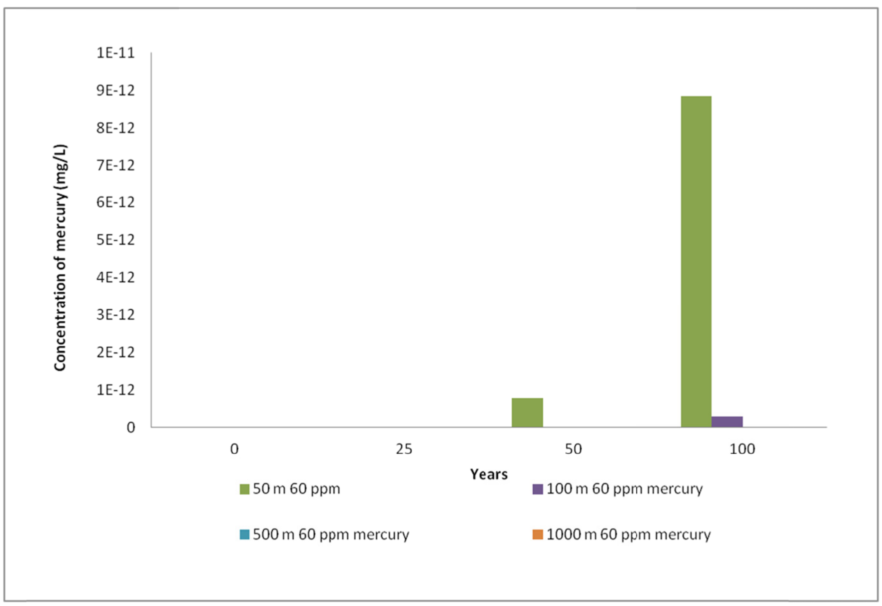

Figure 65 Comparison 95th concentration for all distance to the well (60 ppm)

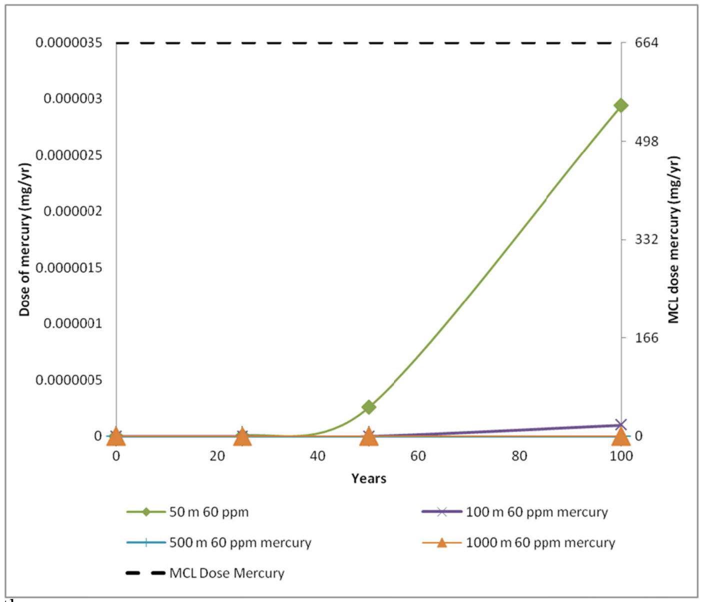

Figure 66 95th dose of mercury for different distance to the well (60 ppm)

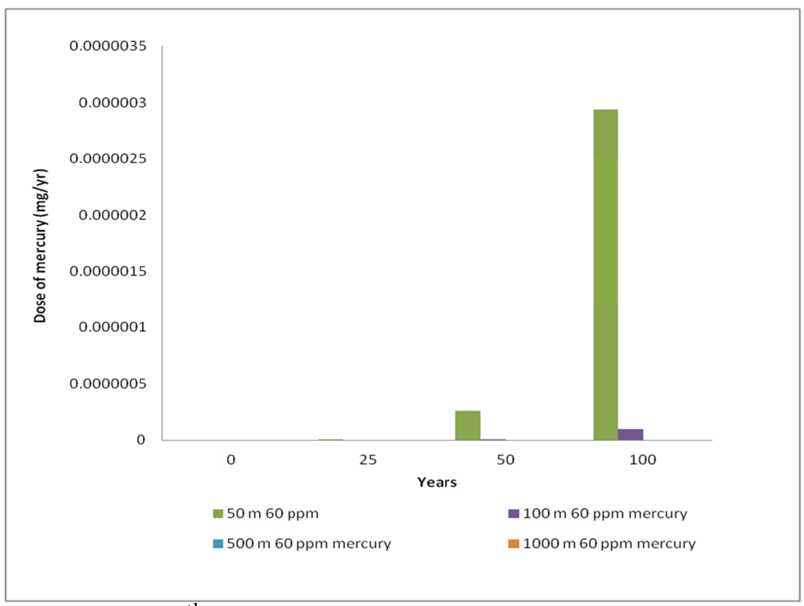

Figure 67 Comparison 95th dose of mercury for all distance to the well (60 ppm)

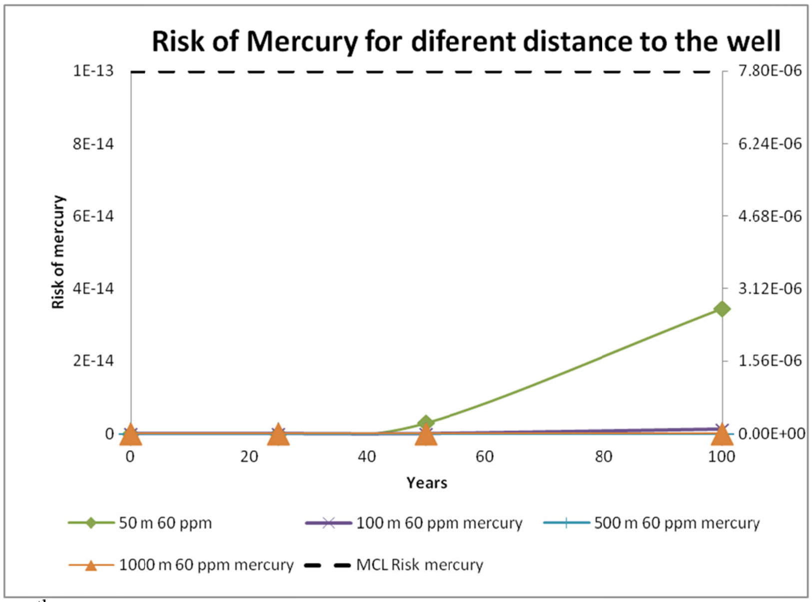

Figure 68 95th percentile risk of mercury for different distance to the well (60 ppm)

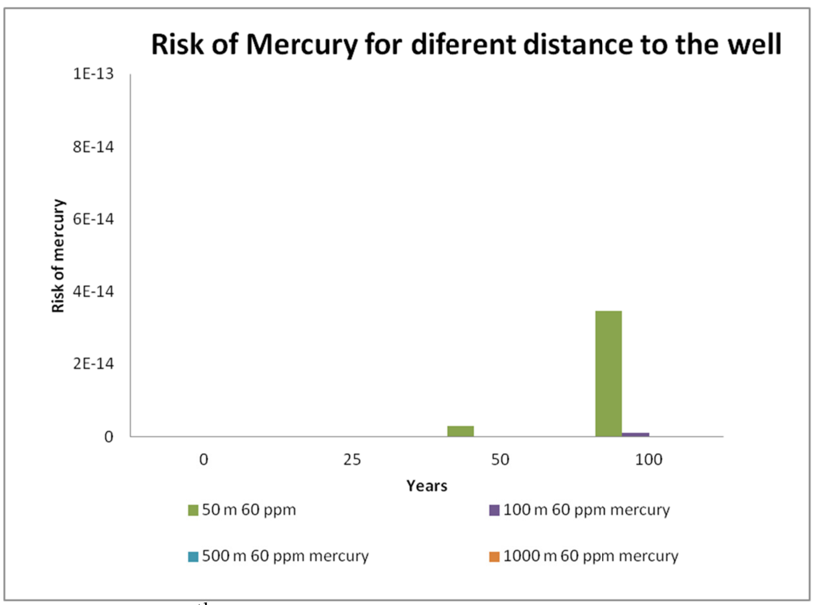

Figure 69 Comparison 95th risk of mercury for all distance to the well (60 ppm)

Table 51 shows a summary of the output for concentration, dose, and risk of mercury of deterministic simulations performed for different distances to a drinking well. The initial concentration of mercury in the repository was increased with a ratio of 1:60 (1 mg/kg (1 ppm) to 60 mg/kg (60 ppm)). The distances for the simulation were 50, 100, 500, and 1,000 meters. For all of these concentrations of mercury the values do not exceed the EPA MCL and do not represent a potential hazard.

Table 51 Summary of simulations for different distances to the well (60 ppm)

Type of Simulation (100 years)	Concentration of mercury 95th (mg/L	Dose of mercury 95th (mg/yr)	Risk mercury 95th (Dimensionless)
50 meter distance	$8.84x10^{-12}$	$2.94x10^{-6}$	$3.45x10^{-14}$
100 meter distance	$3.03x10^{-13}$	$1.01x10^{-7}$	$1.18x10^{-15}$
500 meter distance	$1.83x10^{-25}$	$6.08x10^{-20}$	$7.14x10^{-28}$
1000 meter distance	0	0	0

A deterministic simulation was done for the solubility of mercury (1.47 mol/l) using order of magnitude from 10^{-2} to 10^{2} to evaluate if a change in solubility produces a significant variation in the output of concentration of mercury.

Figure 70 shows the 95^{th} percentile of concentration of mercury for different solubilities. The graph indicates that there is not a change in concentration due to changes in order of magnitude of solubility.

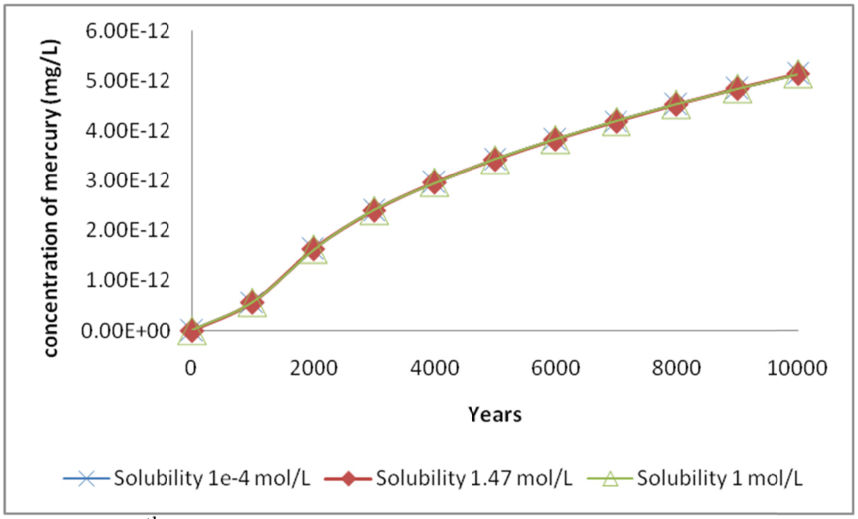

Figure 70 95th concentration of mercury for different solubilities

A deterministic simulation was performed for Q of water (m^3/yr), since Q is the quantity of water to be discharge and is the result of multiply the area (m^2) with the hydraulic conductivity (m/yr). Hydraulic conductivity (K) has a unit of velocity, its value is a function of both porous medium and the fluid. A Q of water describes flow through a porous medium (LaGrega et al., 2001) since the area is a constant value. An increase of the value of hydraulic conductivity means an increase of the value of Q of water, therefore a change of the of value of hydraulic conductivity is a change of Q of water. A series of simulations were performed for selected orders of magnitude of Q of water from 10^2 to 10^4 to evaluate how the variation of Q affects the output, the concentration of mercury. Figure 71 shows the 95th percentile of concentration of mercury for variation of Q of water from one m^3/yr until order of magnitude of 10^4.

A Q4 of water of $1x10^4$ m^3/yr, has the highest value of concentration of mercury $6.54x10^{-11}$ mg/L. The graph indicates that the higher the value of Q of water, the greater the concentration of mercury. Therefore, given that the Q of water depends on the value of the hydraulic conductivity and the area is a constant, when the hydraulic conductivity increases, the concentration of mercury also increases. The ratio between concentrations of mercury from Q4 and Q1 is approximately 13. Both values of concentration of mercury for Q1 and Q4 are well below the MCL limits, which is 0.002 mg/L.

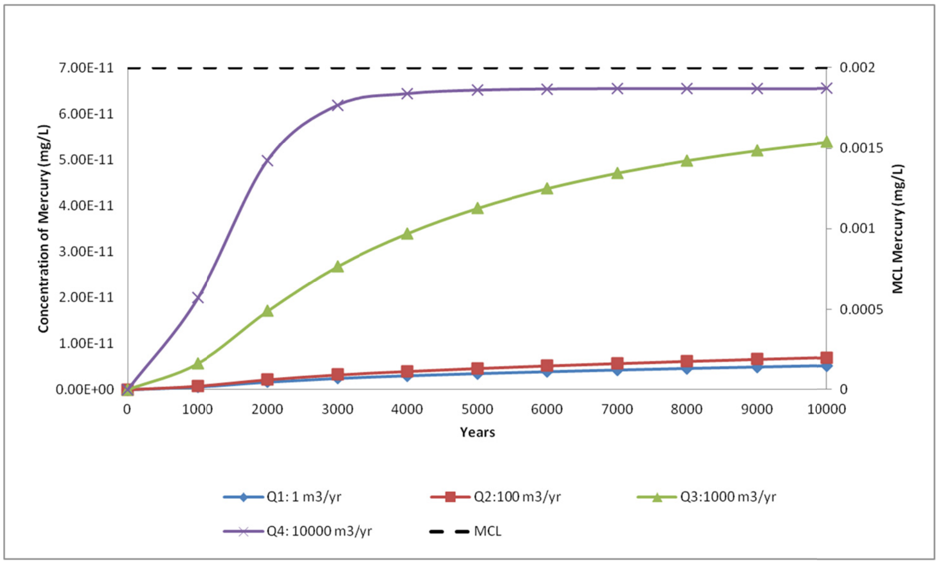

Figure 71 Deterministic simulations for Q of water for 1 ppm

Partition coefficient values represent metal partitioning between the solid phase of waste and its associated with leachate. Partition coefficient obtained from literature data are subject to numerous sources of uncertainty (Allison & Allison, 2005). Therefore, a deterministic simulation set was completed using an order of magnitude for the current values of partition coefficients in the source, and the unsaturated and saturated zones to evaluate their impact in the concentration of mercury.

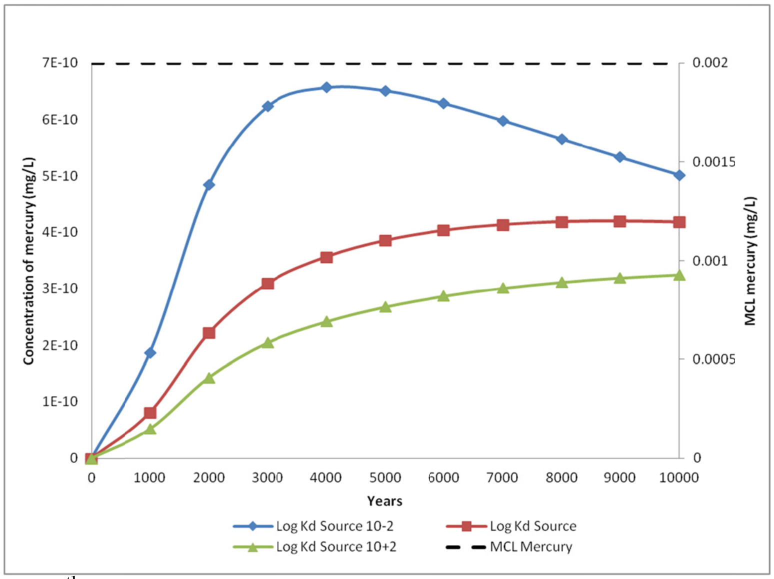

Figure 72 95th concentration for log Kd source different order of magnitude

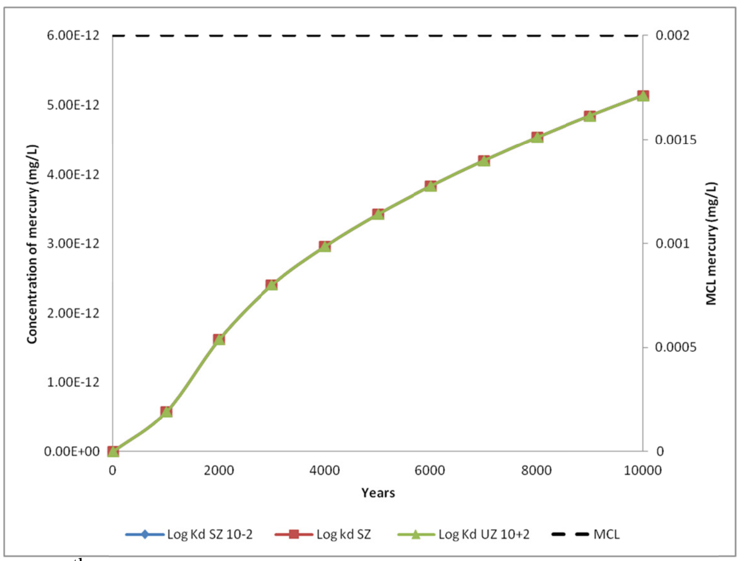

Figure 73 95th concentration for log Kd SZ different order of magnitude

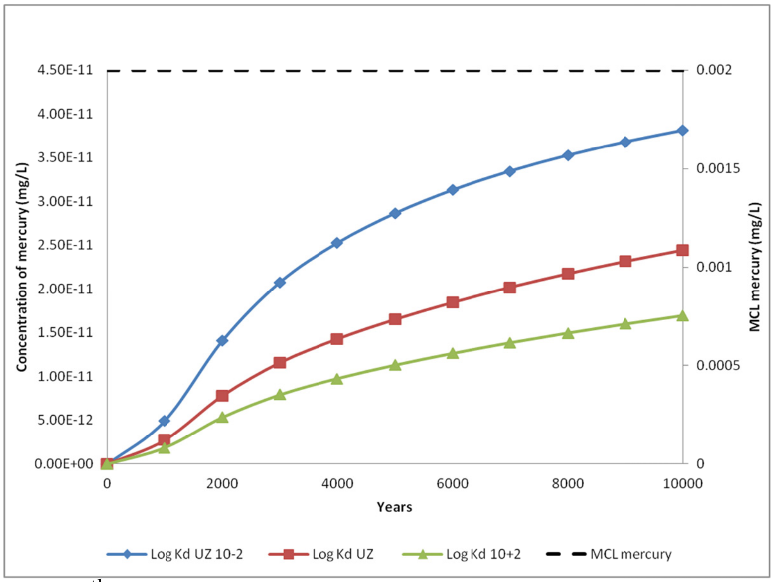

Figure 74 95th concentration for log Kd UZ different order of magnitude

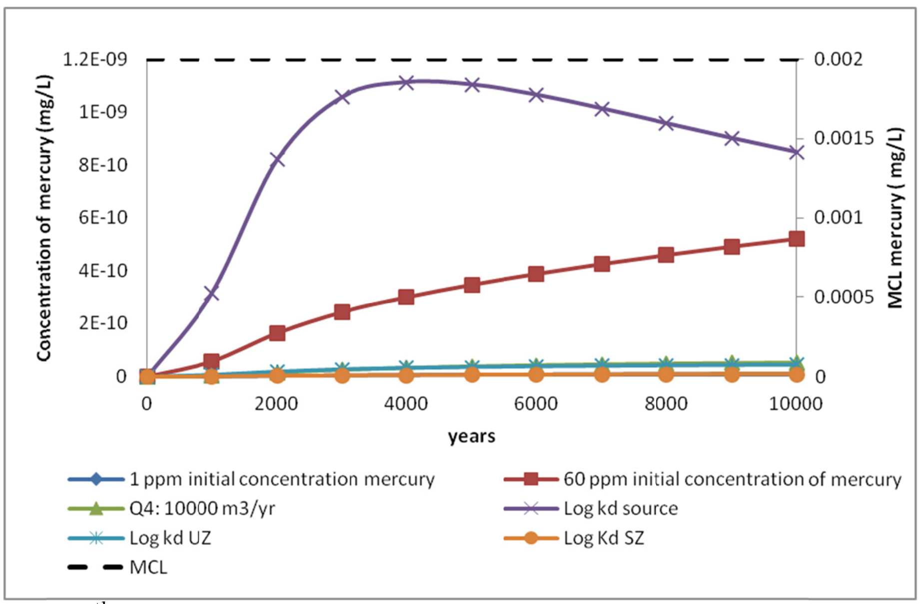

Figure 75 95th concentration for all simulations stochastic porosity (UZ and source)

Table 52 shows the summary of the probabilistic simulations for the stochastic porosity source and unsaturated zone, these simulations were performed using selected deterministic order of magnitude values of paremeters such as Q of water, log Kd source, initial concentration of mercury in the dispository. All the simulations, with the expecion of Kd for the saturated zone, have an output concentration of mercury higher that the initial simulation. All values for concentration, dose, and risk are below the EPA MCL limits.

Table 52 Summary simulations for stochastic porosity (Source and UZ)

Simulation parameter	Value	Units	Conc. 95th (mg/L)	Dose 95th (mg/yr)	Risk 95th
Initial concentration of mercury (1 ppm)	1	mg/kg^3	$8.70x10^{-12}$	$2.89x10^{-6}$	$3.77x10^{-10}$
Initial concentration of mercury (60 ppm)	60	mg/kg^3	$5.22x10^{-10}$	$1.73x10^{-4}$	$2.26x10^{-8}$
Q of water Order of magnitude 10^2	10000	m^3/yr	$5.38x10^{-11}$	$1.79x10^{-5}$	$2.33x10^{-9}$
Log Kd source[1]	Log(5)	mg/L	$8.50x10^{-10}$	$2.82x10^{-4}$	$3.68x10^{-8}$
Log Kd Unsaturated zone	Log(0.41)	mg/L	$4.44x10^{-11}$	$1.47x10^{-5}$	$1.92x10^{-9}$
Log Kd Saturated zone	Log(1)	mg/L	$8.70x10^{-12}$	$2.89x10^{-6}$	$3.77x10^{-10}$
[1].The maximum value was $1.11x10^{-9}$ for a time of 4,000 years					

Table 53 shows the results of the sensitivity analysis performed for the stochastic parameter porosity source and unsaturated zone. These changes of values of the selected deterministic input parameters, have an impact in the output concentrations of mercury but the importance of stochastic porosity source and unsaturated zone is keeping constant.

Table 53 Sensitivity analysis for stochastic Porosity in the UZ for all simulations

Type of Simulation	Coefficient of determination	Corr. Coeff.	SRC	Partial coeff.	Importance Measure
1 ppm	0.99	1	1	1	0.81
60 ppm	0.99	0.99	0.99	-0.99	0.80
Q4:10000 m^3/yr	0.99	-1	-1	-1	0.80
Log Kd Source	0.99	0.99	0.99	0.99	0.80
Log Kd UZ	0.92	0.96	0.96	0.96	0.78
Log Kd SZ	0.99	0.99	0.99	0.99	0.80

ASSUMPTIONS AND LIMITATIONS

For model simulations, Hg transport was assumed non-reactive and assumptions were based on previous studies that indicate that Hg is not physically or chemical reactive in unsaturated porous medium and have limited water solubility. Results from the study presented herein strongly establish the need to confirm the extent of the validity of all assumptions, which requires further data collection and analyses. Future field and modeling work to address the effects and impacts of, for instance, hydraulic conductivity among all other analyzed variables, especially within the source domain and unsaturated zone, should improve the accuracy or discrepancy between predicted and observed Hg concentrations in contaminated ground water. The effects of hydraulic conductivity were not directly addressed in the model, because the GOLDSIM CT module does not have hydraulic conductivity as an input parameter; instead ranges of water flow, which is linearly related to hydraulic conductivity in Darcy's Law, were used as indirect indicators of the effect of hydraulic conductivity.

Although the PRA approach offers advantages over the common point estimate approach in risk assessment, the use of PRA requires additional information on the probability of the variables or parameters of interest, which requires much more data; it also adds some level of complexity to the communication experience between experts and public. In other words, although quantitative risk estimates may be quite informative, they also may be more complext to describe and justify and may not be well received by the public, which may expect much certainty (Slovic, et al. 1979).

Overall, it is important to keep in mind that the main purpose of probabilistic analysis is mostly "screening", providing a general and semiqualitative insight into release pathway characteristics as function of relevant transport parameters, but not to establish detailed facility performance scenarios (Hiergessel & Taylor, 2008).

CONCLUSIONS

To remedy and contain the contamination of mercury in the surroundings areas of Y-12 NSC, a new containment system has been proposed. The EMWMF is the designated area to host the new containment system. To comply with the regulatory standards of the US Department of Energy, the probabilistic assessment of this study was conducted to aid in the evaluation of the potential risk impacts to the public and the environment. The contaminant transport and release from the mercury containment system in Bear Creek Valley to a reference drinking water well, within the surroundings of the proposed containment, was then analyzed using the GOLDSIM contaminant transport module; GOLDSIM can simulate one-dimension advection-dispersion of contaminants in the groundwater.

The model used the Monte Carlo method with PDF distributions of selected soil and hydrological parameters to create probabilistic time series for concentration, dose, and risk of mercury. The model simulated one-dimensional release of mercury from the containment system into the groundwater flow unsaturated and saturated zones to the reference drinking water well. The main purpose was to evaluate how the uncertainty in soil and hydrological parameters, such as infiltration rate, porosity, dry bulk density, and partition coefficient can influence the concentration of mercury in the drinking water well over a period of 10,000 years. Estimates of the concentration of mercury in that drinking water well, for all the simulations, were well below the EPA MCL, which is 0.002 mg/L.

In the simulations, the porosity of the source domain and unsaturated zone presented the highest uncertainty variance, that is 58%, at a 95th percentile for the mercury concentration of 8.7×10^{-12} mg/L. The sensitivity analysis shows that among all the evaluated parameters with the GOLDSIM CT extension, mercury concentration and risk estimates are most sensitive to the porosity of the source domain and unsaturated zone. That sensitivity does linearly extend to the retardation factor.

In general, contaminated groundwater with mercury is not expected to be a hazard, at ground water wells located within one meter from the boundary of the facility (this location is this study's conservative assumption) for a period of time between 100 to 10,000 years. Estimates from a number of deterministic simulations, for different distances to well GW-904 (i.e., 50, 100, 500 and 1000 meters) and for a 100-year period, also indicated that the presence of mercury in the groundwater decreases significantly at a distance of 50 meters. In addition, simulations that were made to assess the effect of best estimated ranges of mercury release concentrations at the source, mercury solubility, groundwater flow rates (when as an indirect indicator of hydraulic conductivity), partitioning coefficient (for mercury Hg^{+2}, the species considered in this study) and porosity in the source domain and unsaturated zone, indicate that changes in concentration and risk should occur over the ranges of parameter values, but should not exceed regulatory limits (of course, under the assumptions of this study).

DISCLAIMER:

This study was prepared to comply with partial requirements of the Master of Science degree program in Environmental Engineering at Florida International University. The author of this thesis, Professor Fuentes, the Committee Members and Florida International University:

1. Do not make any warranty or representation, expressed or implied, with respect to the accuracy and completeness of the information contained in this study.
2. Do not warrant that the use of any information, method or process described in this study may not infringe on privately owned rights.
3. Do not assume any liabilities with respect to the use of or for damages resulting from the use of any information, method or process described in this study.

This study does not reflect the official views or policies of any participating organizations. It was completed as a preliminary literature-based estimation of possible technologies and was based on assumptions, due to lack of access to the site and resources, which need to be carefully acknowledged and properly addressed in any further phase.

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Table 54 Random number used for fitting infiltration rate distributions

Sample	Value	Sample	Value	Sample	Value	Value	Value
1	0.35	15	0.77	29	0.77	43	0.65
2	0.85	16	0.38	30	0.35	44	0.60
3	0.57	17	0.73	31	0.23	45	0.45
4	0.31	18	0.79	32	0.54	46	0.77
5	0.63	19	0.22	33	0.28	47	0.80
6	0.64	20	0.70	34	0.93	48	0.50
7	0.66	21	0.53	35	0.83	49	0.57
8	0.95	22	0.49	36	0.62	50	0.76
9	0.81	23	0.73	37	0.59		
10	0.51	24	0.79	38	0.69		
11	0.39	25	0.69	39	0.61		
12	0.70	26	0.45	40	0.82		
13	0.28	27	0.44	41	0.42		
14	0.71	28	0.65	42	0.84		

Table 55 Values for top position slope for dry bulk density UZ

Site	Dry bulk density	Site	Dry bulk density
7.2.11	1.26	wb4.11	0.87
7.2.12	1.05	wb4.12	1.17
7.2.13	1.04	wb4.21	1.04
7.2.21	1.18	wb4.22	0.7
7.2.22	0.85	bc1.11	0.9
7.2.23	1.19	bc1.12	1.07
7.4.11	1.17	bc1.13	1.12
7.4.12	1.01	bc1.21	1.05
7.4.21	1.12	bc1.22	1.18
7.4.22	1.13	bc1.23	1.01
wb1.11	1.03		
wb2.21	1.27		
wb2.21	1.1		

Table 56 Values for bottom position slope for dry bulk density SZ

Site	Dry bulk density	Site	Dry bulk density
C12.14	0.96	7.2.14	1.03
C12.15	1.17	7.2.21	1.12
C12.24	1.13	7.2.22	1.06
C12.25	0.96	7.2.23	1.15
7.1.11	0.92	7.2.24	1.27
7.1.12	1.18	bc4.21	1
7.1.13	1.29	bc4.11	1.28

Table 57 Random numbers for fitting porosity UZ distributions

Sample	Value	Sample	Value
1	0.66	11	0.94
2	0.51	12	0.2
3	0.03	13	0.6
4	0.23	14	0.18
5	0.88	15	0.12
6	0.13	16	0.13
7	0.54	17	0.71
8	0.19	18	0.64
9	0.86	19	0.55
10	0.7	20	0.94

Table 58 Random number for fitting porosity SZ distributions

Sample	number	Sample	number
1	0.16	11	0.1
2	0.16	12	0.1
3	0.02	13	0.21
4	0.19	14	0.11
5	0.1	15	0.16
6	0.15	16	0.25
7	0.23	17	0.12
8	0.08	18	0.09
9	0.16	19	0.03
10	0.02	20	0.04

Table 59 Random number for fitting Kd distributions containment system

Sample	Number	Sample	Number
1	451	11	317
2	670	12	280
3	301	13	549
4	363	14	609
5	678	15	327
6	511	16	617
7	624	17	601
8	288	18	461
9	645	19	202
10	468	20	413

Table 60 Random number for fitting Kd distributions unsaturated zone

Sample	Number	Sample	Number
1	55	11	47
2	100	12	47
3	95	13	42
4	93	14	63
5	65	15	75
6	71	16	29
7	46	17	81
8	87	18	92
9	76	19	26
10	67	20	23

Table 61 Random number for fitting Kd distributions saturated zone

Sample	Number	Sample	Number
1	169	11	180
2	119	12	100
3	117	13	167
4	101	14	100
5	145	15	151
6	95	16	95
7	93	17	90
8	158	18	111
9	140	19	142
10	193	20	196

Table 62 Mercury concentrations deterministic simulation 1000 realizations

Time (yr)	Mean	S.D.	Least Result	5%	95%	Greatest Result
0	0	0	0	0	0	0
1000	5.73E-13	0	5.73E-13	5.73E-13	5.73E-13	5.73E-13
2000	1.62E-12	0	1.62E-12	1.62E-12	1.62E-12	1.62E-12
3000	2.40E-12	0	2.40E-12	2.40E-12	2.40E-12	2.40E-12
4000	2.96E-12	0	2.96E-12	2.96E-12	2.96E-12	2.96E-12
5000	3.43E-12	0	3.43E-12	3.43E-12	3.43E-12	3.43E-12
6000	3.83E-12	0	3.83E-12	3.83E-12	3.83E-12	3.83E-12
7000	4.20E-12	0	4.20E-12	4.20E-12	4.20E-12	4.20E-12
8000	4.53E-12	0	4.53E-12	4.53E-12	4.53E-12	4.53E-12
9000	4.85E-12	0	4.85E-12	4.85E-12	4.85E-12	4.85E-12
10000	5.14E-12	0	5.14E-12	5.14E-12	5.14E-12	5.14E-12

Table 63 Mercury concentrations deterministic simulation 100 realizations

Time (yr)	Mean	S.D.	Least Result	5%	95%	Greatest Result
0	0	0	0	0	0	0
1000	5.73E-13	0	5.73E-13	5.73E-13	5.73E-13	5.73E-13
2000	1.62E-12	0	1.62E-12	1.62E-12	1.62E-12	1.62E-12
3000	2.40E-12	0	2.40E-12	2.40E-12	2.40E-12	2.40E-12
4000	2.96E-12	0	2.96E-12	2.96E-12	2.96E-12	2.96E-12
5000	3.43E-12	0	3.43E-12	3.43E-12	3.43E-12	3.43E-12
6000	3.83E-12	0	3.83E-12	3.83E-12	3.83E-12	3.83E-12
7000	4.20E-12	0	4.20E-12	4.20E-12	4.20E-12	4.20E-12
8000	4.53E-12	0	4.53E-12	4.53E-12	4.53E-12	4.53E-12
9000	4.85E-12	0	4.85E-12	4.85E-12	4.85E-12	4.85E-12
10000	5.14E-1	0	5.14E-12	5.14E-12	5.14E-12	5.14E-12