Assessment of Migration of Radionuclides from A Hypothetical Radioactive Waste Repository Sited in the Schist Rock of the Accra Plains

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Abstract:- **The Geosphere of the Accra Plains, made up of the very low permeable Togo and Dahomeyan rocks is a potential site to host a disposal facility for disused radioactive sources generated in Ghana. The geosphere constitutes a natural barrier for the possible leakage of radionuclides from the proposed disposal facility. This study assessed the migration of radioactive contaminant through the geosphere to the biosphere, from a hypothetical radioactive waste disposal facility sited in the Accra Plains. Lithologies of thirty-one (31) boreholes in the Plains were compiled and compared to determine a representative lithology of the Plains to develop the conceptual geological model. The geosphere between the near field of the disposal facility and an abstraction borehole situated down the hydraulic gradient from the disposal facility was discretised into sixty (60) compartments consisting of a rock matrix with a single planar fracture in water saturated porous rock. Transport in the fracture is assumed to obey an advection-diffusion equation, while molecular diffusion is considered the dominant mechanism of transport in porous matrix. The processes that have the potential to move contaminants around the system were represented as transfers between those compartments. This was implemented in the AMBER software. Radiation exposure of humans is an estimated effective dose to an adult from ingestion of well water from an abstraction borehole. The study considered abstraction boreholes located 100m, 500m, 2km and 5km from the disposal facility. The predicted results for liquid releases in terms of the calculated peak total dose in all cases were significantly lower than the dose constraint of 0.3mSv/y. The results thus indicate that the Accra Plains is capable of hosting a disposal facility for Ghana's disused radioactive sources.**

Keywords: Radioactive Waste, Contaminant, Compartment, Transport, Geosphere

INTRODUCTION

An international consensus within the nuclear industry and among governments exists that, the emplacement of radioactive waste in geological repository is the best long term radioactive waste management option that is currently available (E.T. Glover, 2012). Geological repository is a facility for the disposal of radioactive waste located underground, generally from a few tens of meters to some hundreds of meters below the surface in a stable host rock. This is intended to provide long term isolation of radionuclides from the accessible environment (IAEA, 2003).

The use of radioactive materials in Ghana dates back in the 1950s in the then University College of Gold Coast, now the University of Ghana (Glover E. T. and Fletcher, J. J., 2000). Radioactive sources have since been in use in various sectors of the Ghanaian economy viz.: medicine, agriculture, industry, research and academics. Some of these applications include; Nuclear medical centers (for radiotherapy, diagnostics, equipment sterilization); Food processing facility (for food preservation & shelf-life extension through irradiation*);* as nuclear gauges in the mining, road construction, breweries, oil and gas exploration and production industries besides other petrochemical industries, smoke detectors as well as in R&D laboratories (for calibration sources, gamma and neutron irradiators).

As a result of the applications of radioactive materials, radioactive waste materials are generated. The radioactive nature of the waste material makes it potentially dangerous and hence presents a potential risk to human health and the environment. These waste materials must be specifically managed safely to avert any associated risks now and in the future. It is required by law and by principle that any society or organization which benefited from the use of the radioactive materials should act in a responsible manner with respect to mankind and the environment so as not to put undue burden on future generations.

Though efforts can be made to minimise the generation of radioactive waste materials by storing the waste safely, there will always remain some long lived waste materials which inevitably have to be disposed of in underground repositories. Eventually, high-level radioactive wastes will be deposited in deep geological repositories in crystalline rocks in many countries (Li and Chiou, 1993; Neretnieks, 1993; Gylling et al., 1998; Batoul M., 2016). Geological disposal is a model of "concentrate and contain" method whereby containment is achieved so as to have a negligible effect on the biosphere for thousands of years, NEA (1999). Geological disposal is also a multi-barrier approach where engineered barriers and geological environment surrounding the disposed radioactive waste work together to provide variety of security features, which control all possible radionuclide releases and their movement through the host rock (Falck and Nilsson 2009, Claudia S., 2013)

One of the basic problems of the final disposal of nuclear waste is how to ensure isolation of the waste from the biosphere for extended time spans as some have radionuclides with long half-life (e.g. Ra-226 with half-life of 1600 yrs.). Moving groundwater and the prevailing chemical conditions can induce degradation of the nuclear waste disposed and transport the released radionuclides to the biosphere through porous and fracture network in the geosphere (Jussila Petri, 2000).

As long term performance needs to be achieved over the long period of disposal, the engineered barriers will undergo degradation leading to the migration of radionuclides into the groundwater and subsequently transported to the biosphere through the geosphere.

The Geosphere of the Accra Plains is a potential site to host such a repository. Dawood et al., 2012, assessed a site in the Accra Plains earmarked for a radioactive waste disposal facility by means of seismic refraction survey.

Glover E. T. (2013), worked on the hydrogeochemical characterization of the rocks in the Accra Plains for a radioactive waste repository. The study concluded that the hydrogeochemical conditions of the rocks at Okwenya and the foothills of the Krobo Mountains within the Plains were found to be favourable for a radioactive waste repository. This was based on the fact that the mafic gneiss at Okwenya and the foothills of the Krobo Mountains contain redox-sensitive minerals. These minerals are magnetite, biotite, chlorite, and amphiboles which can react with groundwater to provide reducing conditions in the aquifer required for a repository.

Yeboah et al., (2014) modelled the migration of radionuclide in a proposed radioactive waste borehole disposal site in the Accra Plains numerically. This study established that radionuclides that will be released from the radioactive waste repository will migrate through preferential pathways within the fracture medium and also, differences in hydraulic conductivities attributable to heterogeneity will affect the direction of flow

The geosphere within which the proposed radioactive waste repository is to be sited serves as natural barrier to isolate radionuclides from the environment. This geosphere however, has fractures and pores through which released radionuclides can migrate to the environment. There is therefore the need to monitor the released contaminant as it migrates through the geosphere so as to mitigate its effects on humans and the environment.

STUDY AREA

The Accra Plains also referred to as the Eastern Lowlands/Coastal Plains is located on the eastern part of the Akwapim Range and underlain by gentle, mature topography which slightly slopes towards the ocean (Muff, R. and Efa, E., 2006). It is bounded on the west and the northwest by the Akwapim Togo Mountains and in the north to the east by the Volta River and on the south by the Gulf of Guinea (Glover, E.T., 2013).

The Accra Plains has no pronounced topography and rare outcrops but has a pedimented surface and forms a low-lying terrain (Burke, K., 1969, Agbevanu, 2015). The directions of main rivers are recognized as lineaments in aerial photographs due to steeply dipping faults, preferred directions of dike intrusions, joint or shears presenting possible zones of weakness in the rocks (Muff et. al., 2006, Agbevanu 2015)

The Accra Plains lies between latitude $5^0 \, 30^1$ N and $6^0 \, 15^1$ N of the equator, and latitude $0^0 \, 20^1$ W and $0^0 \, 40^1$ E of the Greenwich meridian and is situated on the south-eastern part of Ghana. The Plains approximately covers 7000 km^2 (Kesse, 1985, Glover, 2013).
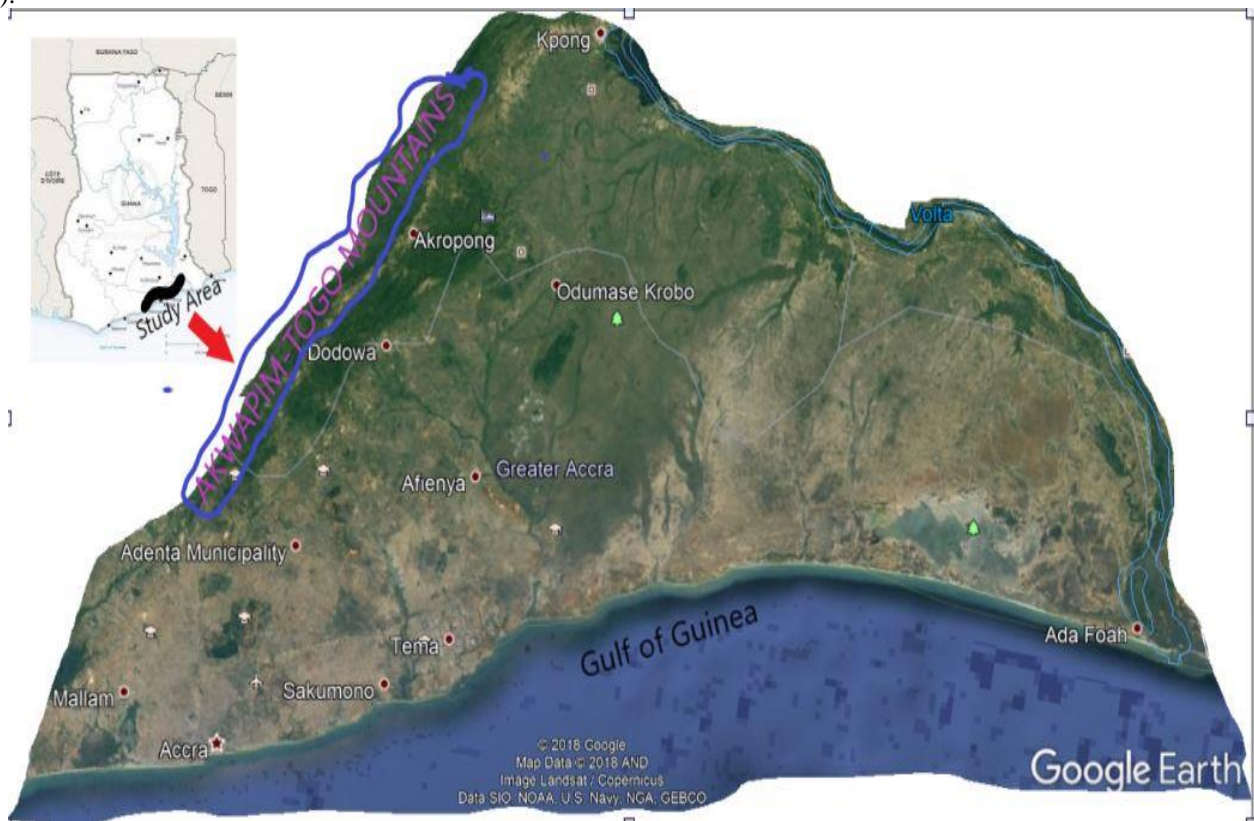

Figure 1: Location Map of the Accra Plains

METHODOLOGY

Lithologies of 31 boreholes in the Accra Plains, Table 1, were compiled to come out with the conceptual geological model.

Table 1: Summary of lithology of 31 boreholes in the Accra Plains

Borehole IDs	Laterite	Clay	Quartzite	Phyllite	Mixed Materials	Schist	Weathered Gneiss	Fresh Gneiss
GAEC(BH1&2)	0-4m	4-6m	6-52m	12-19m 37-38m	50-122m	110-150m	---------	---------
Legon (12 boreholes)	0-9m	---------	9-85m		---------	---------	---------	---------
BH1	0-10m	---------	---------	---------	---------	10-80m	---------	---------
BH2	0-5m	---------	---------	---------	---------	10-52m	---------	---------
BH3	0-15m	---------	---------	---------	---------	20-70m	---------	---------
BH4	0-9m	---------	---------	---------	---------	9-70m	---------	---------
BH5	0-5m	---------	---------	---------	---------	10-55m	---------	---------
BH6	0-6m	---------	---------	---------	---------	6-90 m	---------	---------
BH7	0-5m	---------	---------	---------	---------	5-60m	---------	---------
BH8	0-5m	---------	---------	---------	---------	5-60m	---------	---------
BH9	0-4m	---------	4-57m	---------	---------	---------	---------	---------
BH10	0-5m	---------	---------	---------	---------	5-90m	---------	---------
Madina	---------	0-4.2m	---------	---------	---------	---------	4.2-36m	36-60m
Shiashie	---------	0-3m	---------	---------	---------	---------	3-12m	12-150m
Abelempke	---------	0-3m	---------	---------	---------	---------	3-33m	33-80m
CSIR Guest House	---------	0-3m	3-70m	---------	---------	---------	---------	---------
Ghana Standards Authority	0-4m	---------	---------	---------	---------	---------	6-25m	25-141m
Mataheko	10-15m	0-10m	---------	---------	15-45m	---------	---------	---------
Adjiringanor	---------	0-2m	---------	---------	---------	---------	2-4m	4-130m

From the summary of lithology of the 31 boreholes in the Accra plains, the following conceptual geological model (Fig. 3) for the Plains was deduced;
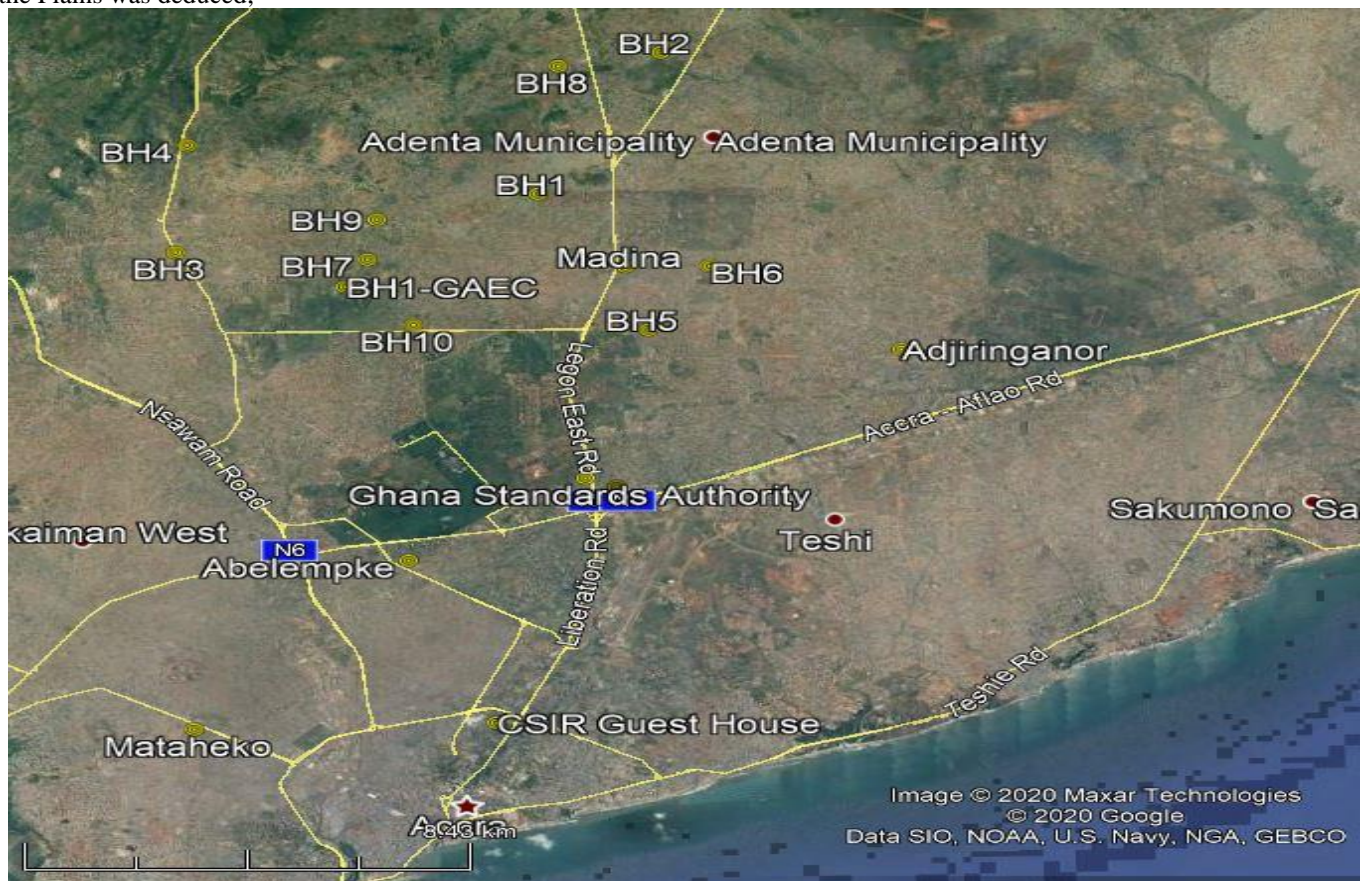

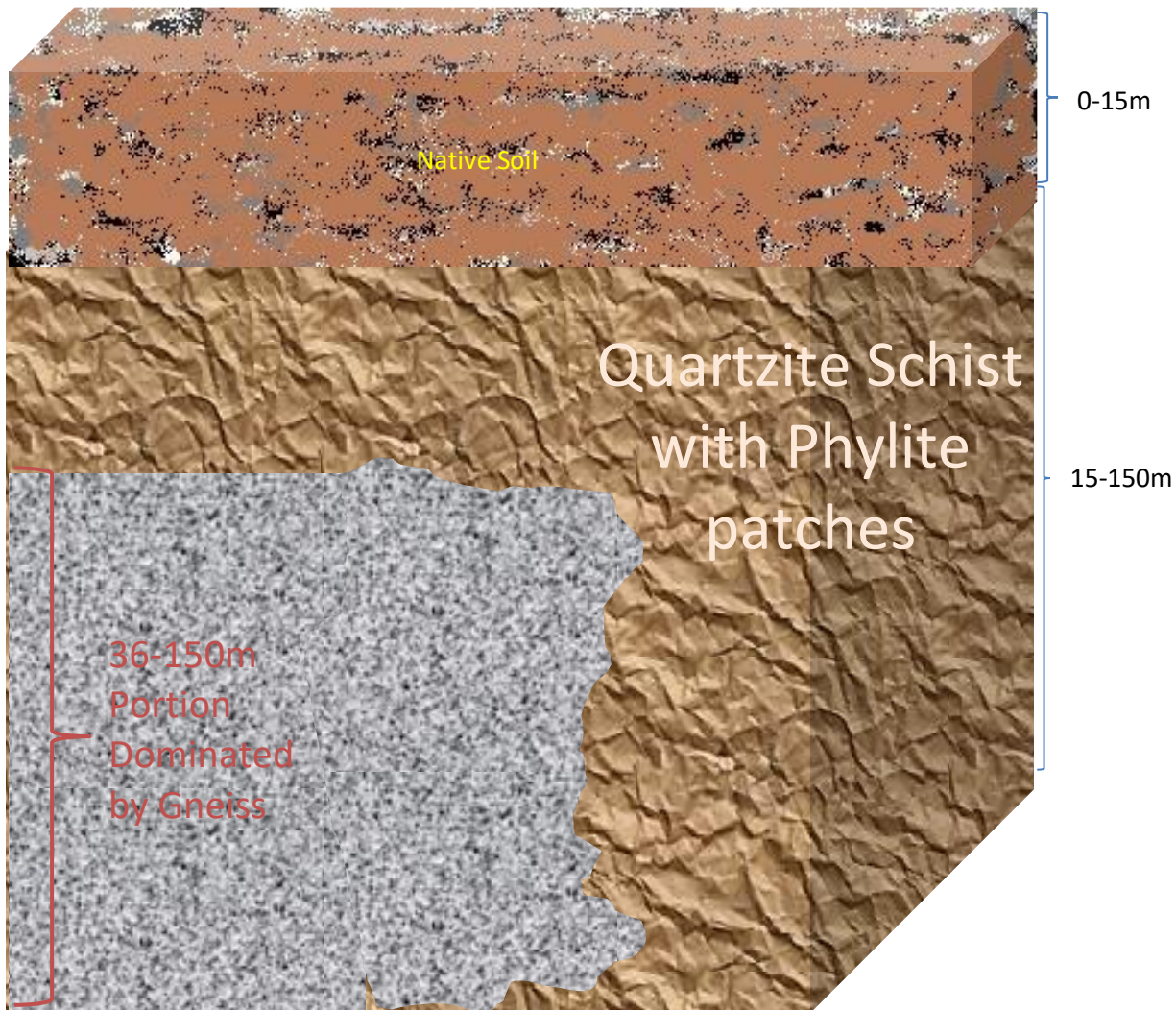Figure 2: Location of Eighteen (18) Boreholes in the Accra Plains

Figure 3: Conceptual Geological Model of the Accra Plains

From the conceptual geological model, the Plains is predominantly underlain by Schist and Gneiss rocks. The properties of Schist rocks, (i.e., hydraulic conductivity, porosity etc.) however were used as the input parameters in the model development.

From the conceptual geological model, the physical modeling domain with the discretised geosphere as illustrated in Fig.4 can be proposed;
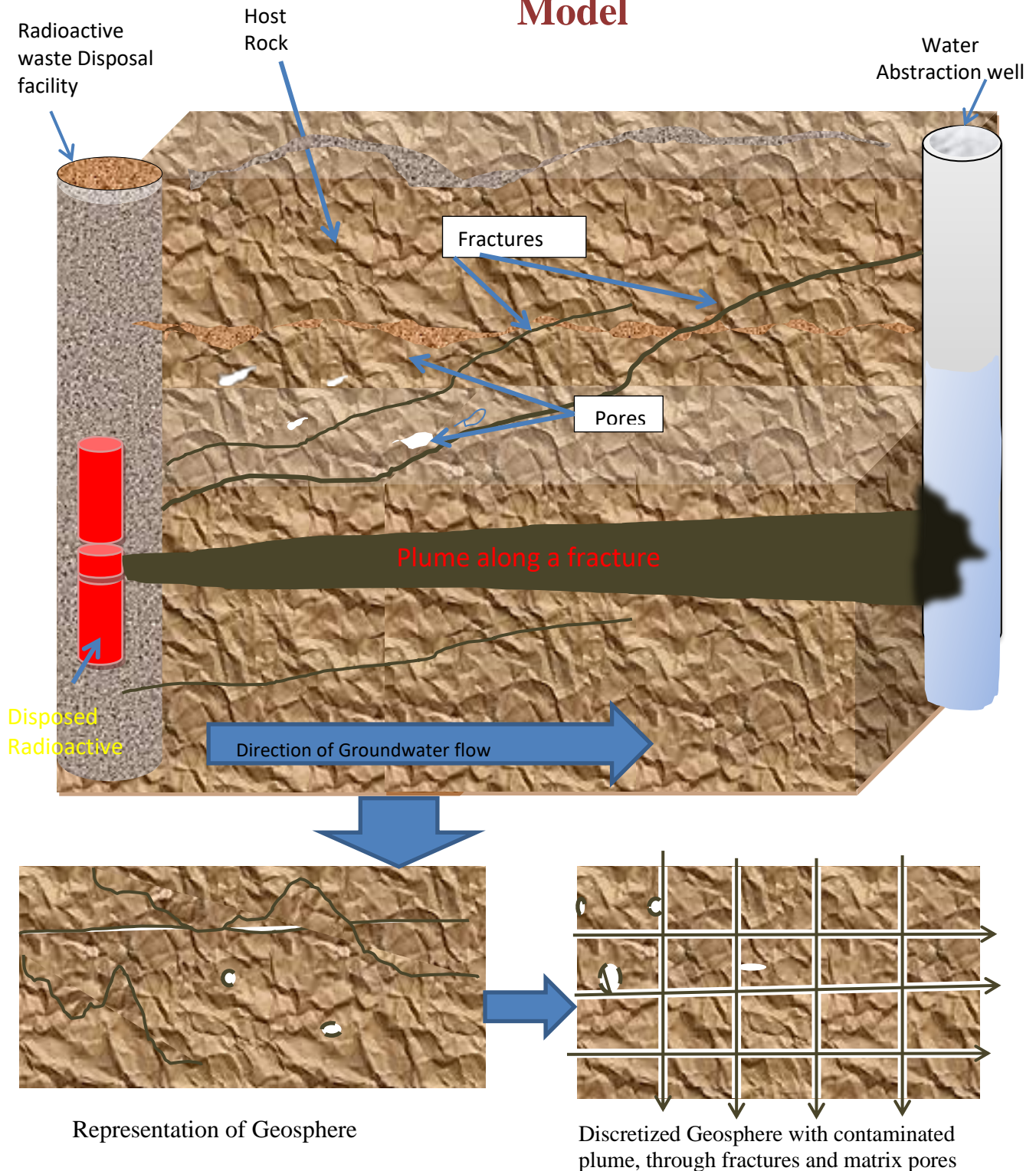
Conceptual Model

Figure 4: Modeling Physical Domain

CONCEPTUAL MODEL

The physical problem being modelled is as follows:

Radionuclides are migrating from a hypothetical radioactive waste repository sited in the Accra Plains. These radionuclides are being transported through the fractures in the geosphere to the biosphere by flowing groundwater through advection, longitudinal hydrodynamic dispersion along the fracture axis, with sorption onto the surface of the fracture. The flow in the fractures is coupled with molecular diffusion from the fracture into the rock matrix, with sorption within the rock matrix, and radioactive decay.

The task is to develop a dual porosity model to assess the fate of the transporting radionuclides in the geosphere to determine the spatial and temporal activity concentration to predict the implications on humans and the environment

The following assumptions were made to develop the problem geometry;

i. The length of the fracture L is far greater than its width, w
ii. The radionuclides in the groundwater being transported in the fracture are well mixed such that the concentration is uniform in any plane perpendicular to the direction of flow.
iii. Transport along the fracture is much faster than transport in the pores of the rock matrix.
iv. Transport in the rock porous matrix is controlled by molecular diffusion
v. The water flow is laminar in the fracture.

In view of assumptions i and ii, the mass transfer along the fracture can be described by one-dimensional rectangular coordinate. Considering assumptions iii and iv, the mass flow in the rock pores can be taken in the direction perpendicular to the fracture plane. Assumption v allows the use of Darcy law, Chambre et al., (1982). Based on this law, it follows that the flow is described by a mean linear speed which carries the dissolved radionuclides by convection. This is diagrammatically shown in Fig. 5;
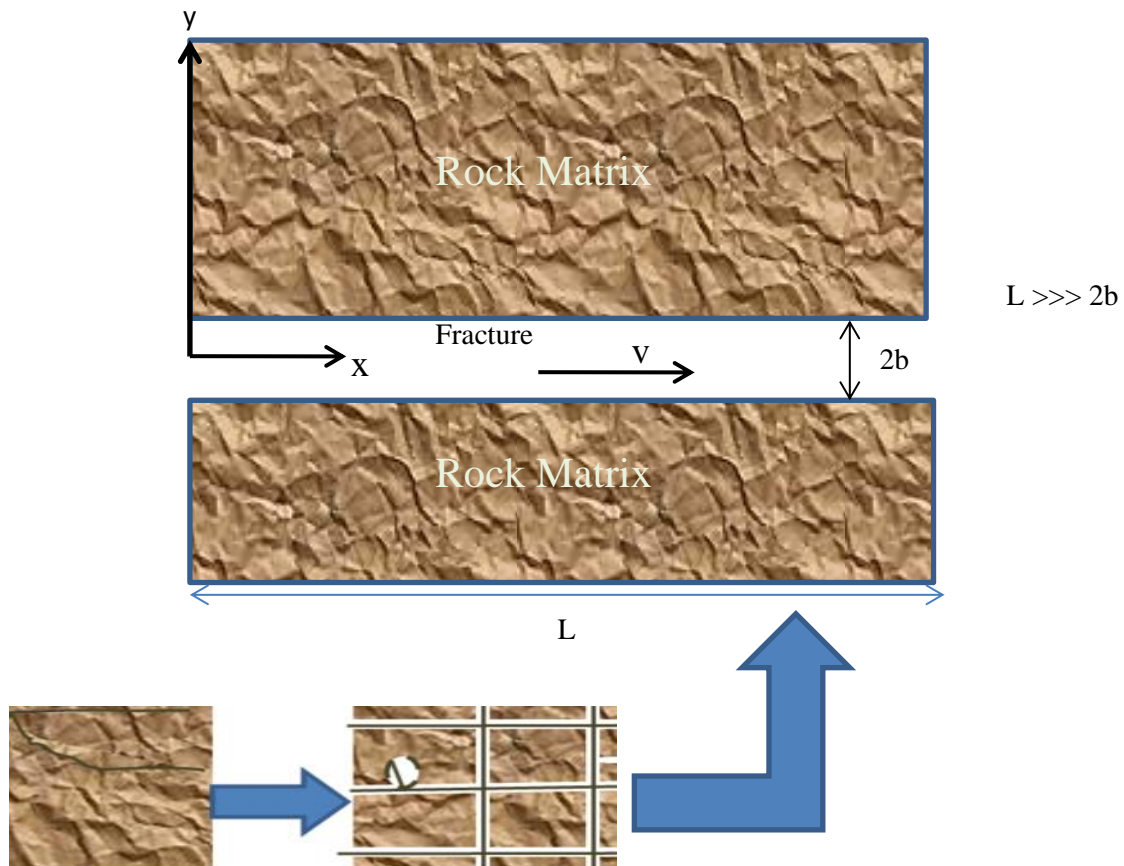

Figure 5: Model geometry with a single fracture

GOVERNING EQUATIONS

Two coupled one-dimensional equations describe the transport process of the decay chain of contaminants containing radionuclides. Based on the aforementioned assumptions, one equation for the fracture representing advective transport and other for the porous matrix representing diffusive transport in the porous matrix can be expressed. The coupling is provided by the continuity of fluxes and concentrations along the interface. The differential equations describing the distribution of a solute of n radionuclide species along the fracture is given by (Tang et.al., 1981, Hedin, 2000);

$$\frac{\partial C_{f,1}(x,t)}{\partial t} + \frac{v}{R_{f,1}}\frac{\partial C_{f,1}(x,t)}{\partial x} - \frac{D_{f,1}}{R_{f,1}}\frac{\partial^2 C_{f,1}(x,t)}{\partial x^2} + \lambda_1 C_{f,1}(x,t) + \frac{q_1(x,t)}{R_{f,1}b} = 0$$

$$\dots\dots\dots\dots\dots\dots\dots\dots$$
$$\dots\dots\dots\dots\dots\dots\dots\dots$$
$$\dots\dots\dots\dots\dots\dots\dots\dots$$

$$\frac{\partial C_{f,n}(x,t)}{\partial t} + \frac{v}{R_{fn}}\frac{\partial C_{f,n}(x,t)}{\partial x} - \frac{D_{f,n}}{R_{fn}}\frac{\partial^2 C_{f,n}(x,t)}{\partial x^2} + \lambda_n C_{f,n}(x,t)$$
$$- \frac{R_{f,n-1}\lambda_{n-1}C_{f,n-1}(x,t)}{R_{f,n}} + \frac{q_n(x,t)}{R_{f,n}b} = 0$$
$$for\ x > 0, t > 0$$

…. (1)

where;

C_f is the fracture solute concentration [M/L^3],

D is the diffusion/dispersion coefficient [L/T],

v is the groundwater velocity in the fracture [L/T],

t is the time [T],

x is the coordinate in the flow direction [L],

R_f is the adsorption coefficient for the fracture rock interface governed by (Freeze and Cherry, 1979)

$R_{f,n} = 1 + \frac{K_{f,n}}{b}$…………. (2)

where $K_{f,n}$ is the surface distribution coefficient of solute per unit area of the unit volume.

$\lambda_n[1/T]$, the radioactive decay constant of radionuclide n is defined by

$\lambda_n = \frac{ln2}{t_{1/2}(n)}$………………… (3)

where $t_{1/2}(n)$ is the half-life of radionuclide n.

The mass loss of the radionuclide n by decay is represented by the fourth term on the left hand side of the nth eqn.(1) whilst the fifth term in the nth equation in eqn.(1) represents the mass gain of radionuclide n by radioactive disintegration of its parent.

The differential equation describing the diffusive transport of the radionuclide chain in the matrix perpendicular to the fracture is given by (Tang et.al., 1981, Hedin, 2000);

$$\frac{\partial C_{m,1}(x,y,t)}{\partial t} - \frac{D_{m,1}}{R_{m,1}}\frac{\partial^2 C_{m,1}(x,y,t)}{\partial y^2} + \lambda_1 C_{m,1}(x,y,t) = 0$$

$$\dots\dots\dots\dots\dots\dots\dots\dots$$
$$\dots\dots\dots\dots\dots\dots\dots\dots$$
$$\dots\dots\dots\dots\dots\dots\dots\dots$$

$$\frac{\partial C_{m,n}(x,y,t)}{\partial t} - \frac{D_{m,n}}{R_{m,n}}\frac{\partial^2 C_{m,n}(x,y,t)}{\partial y^2} + \lambda_n C_{m,n}(x,y,t) - \frac{R_{m,n-1}\lambda_{n-1}C_{m,n-1}(x,y,t)}{R_{m,n}}$$
$$= 0$$
$$for\ y \geq b, x > 0, t > 0$$

…. (4)

where

$C_{m,n}$ is the concentration of radionuclide n in the rock matrix [M/L^3], and
y is the spatial coordinate perpendicular to the fracture axis [L].

$R_{m,n}$ is the retardation coefficient representing the adsorption of the radionuclide n in solution in the rock matrix, defined by (Freeze and Cherry, 1979);
$$R_{m,n} = 1 + \frac{\rho_m K_{m,n}}{\theta}\dots\dots\dots\dots\dots(5)$$
where

$K_{m,n}$ is the distribution coefficient of radionuclide n in solution in the rock matrix [L^3/M],
ρ_m is the density of the rock matrix [M/L^3] and
θ is the porosity of the rock [ ]
$D_{m,n}$[L^2/T] is the effective molecular diffusion coefficient of radionuclide n and is defined as;
$$D_{m,n} = \tau D^*\dots\dots\dots\dots (6)$$

where, τ is the tortuosity of the rock .
The loss term q(x,t) in (1) can be written as
$$q(x,t) = -D_m \frac{\partial C_m}{\partial y}\dots\dots\dots\dots\dots (5),$$
which is substituted into (1) to yield

$$\frac{\partial C_{f,1}(x,t)}{\partial t} + \frac{v}{R_{f,1}}\frac{\partial C_{f,1}(x,t)}{\partial x} - \frac{D_{f,1}}{R_{f,1}}\frac{\partial^2 C_{f,1}(x,t)}{\partial x^2} + \lambda_1 C_{f,1}(x,t) - \frac{D_{m,1}}{R_{f,1}b}\frac{\partial C_m(y,t)}{\partial y}$$
$$= 0$$

$$\dots\dots\dots\dots\dots\dots\dots\dots$$
$$\dots\dots\dots\dots\dots\dots\dots\dots$$
$$\dots\dots\dots\dots\dots\dots\dots\dots$$

$$\frac{\partial C_{f,n}(x,t)}{\partial t} + \frac{v}{R_{fn}}\frac{\partial C_{f,n}(x,t)}{\partial x} - \frac{D_{f,n}}{R_{fn}}\frac{\partial^2 C_{f,n}(x,t)}{\partial x^2} + \lambda_n C_{f,n}(x,t)$$
$$- \frac{R_{f,n-1}\lambda_{n-1}C_{f,n-1}(x,t)}{R_{f,n}} - \frac{D_{m,n}}{R_{f,n}b}\frac{\partial C_{m,n}(y,t)}{\partial y} = 0$$
$$for\ x > 0, t > 0 \qquad \dots\dots\dots\dots (6)$$

With initial and boundary conditions:

$C_{f,n}(x, 0) = 0$……………………. (7)

$C_{f,n}(0, t) = C_0(t)$…………………. (8)

$C_{f,n}(\infty, t) = 0$………………....... (9)

$C_{m,n}(x, y, 0) = 0$…………………. (10)

$C_{m,n}(b, y, t) = C_{f,n}(y, t)$………….. (11) and

$$\frac{\partial C_{m,n}(\infty, x, t)}{\partial_y} = 0 \dots \dots \dots \dots .. (12)$$

This was implemented in the AMBER Software, a compartmental software tool (version 6.2) (Environs QuantiSci and Quintessa Ltd, 2017). AMBER uses a compartment model approach to represent the migration and fate of contaminants in the disposal system. Using the compartment modelling approach, the disposal system was discretised into twenty (20) linear compartments for the fracture and forty (40) compartments representing the rock matrix. This correspond to the components identified in the conceptual model, (Fig 6).
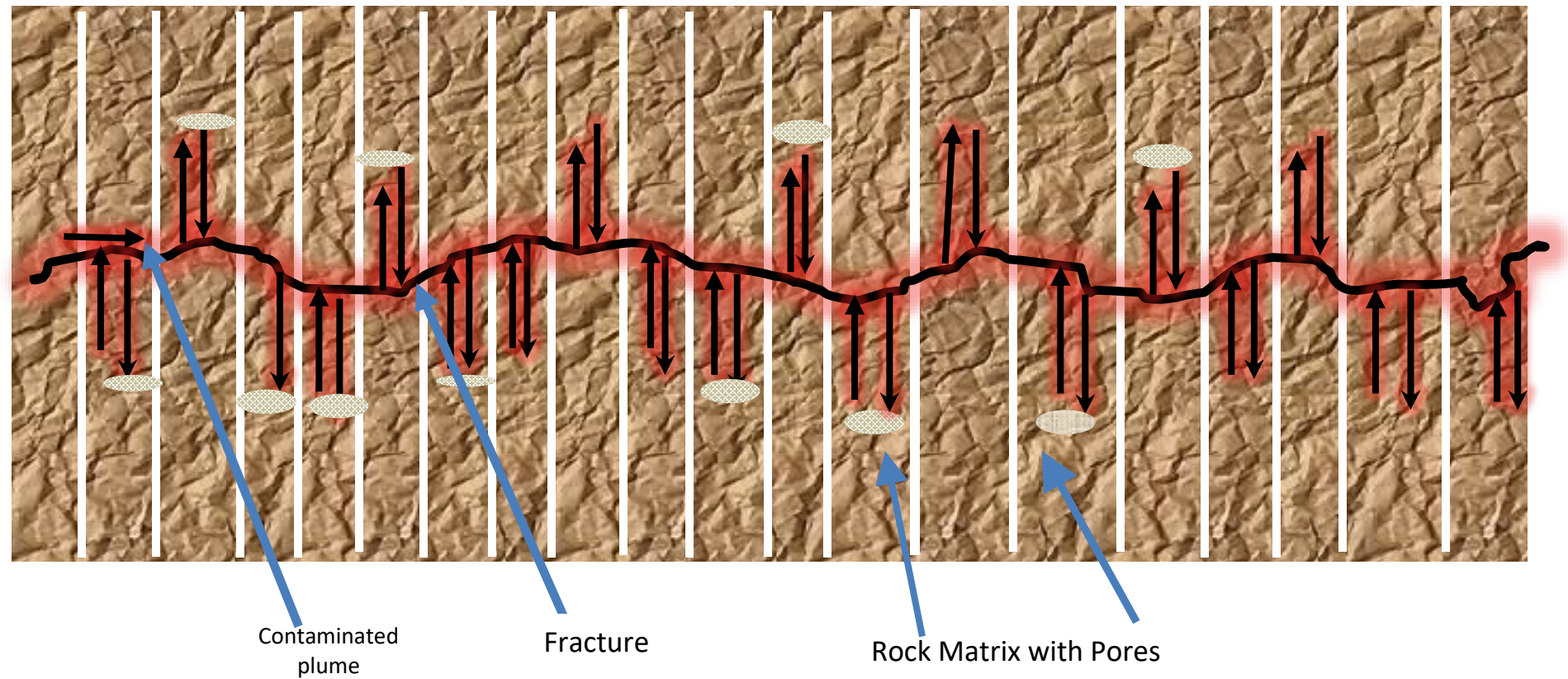
Figure 6: Discretised Modelling domain

It is assumed that either uniform mixing occurs over the timescales of interest, or the distribution of the contaminant within the compartment is not important so that a uniform concentration over the whole compartment can be used either for subsequent transport or for deriving end points of interest. Therefore each compartment should be chosen to represent a system component for which one or other of these assumptions is reasonable.

Secondly, processes resulting in the transfer of contaminants from one compartment (the donor compartment) to another (the receptor compartment) were expressed as transfer coefficients that represent the fraction of the activity in a particular compartment transferred from the donor compartment to the receptor compartment per unit time.

The mathematical model developed in order to assess the distribution of the radionuclide concentration in the geosphere and, consequently, the radionuclide transfer time, is mainly based on the release, migration and transport of radionuclides from the radioactive waste disposed, considering only the decay, adsorption, dispersion and advection processes. Advection and diffusion are along the fracture and diffusion occurs between the fracture and the rock matrix. The mathematical representation of the inter-compartmental transfer processes takes the form of a matrix of transfer coefficients that allows the compartmental amounts to be represented as a set of first order linear differential equations. For the ith compartment, the rate at which the inventory of radionuclides in a compartment changes with time is given by (Quintessa, 2007);

$$\frac{dN_i}{dt} = \left(\sum_{j \neq i} \lambda_{ji} N_j + \lambda_N M_i + S_i(t)\right) - \left(\sum_{j \neq i} \lambda_{ij} N_i + \lambda_N N_i\right) \ldots\ldots\ldots(13)$$

i and j indicate compartments,

N and M are the amounts (Bq) of radionuclides N and M in a compartment (M is the precursor of N in a decay chain).

S(t) is a time dependent external source of radionuclide N (Bq y^{-1}).

Transfer and loss rates are represented by λ;

λ N is the decay constant for radionuclide N (y^{-1}) and

λji and λij are transfer coefficients (y^{-1}) representing the gain and loss of radionuclide N from compartments i and j.

MODEL DEVELOPMENT IN THE AMBER SOFTWARE

The mathematical model was implemented in the AMBER computer code as illustrated in Fig. 7;
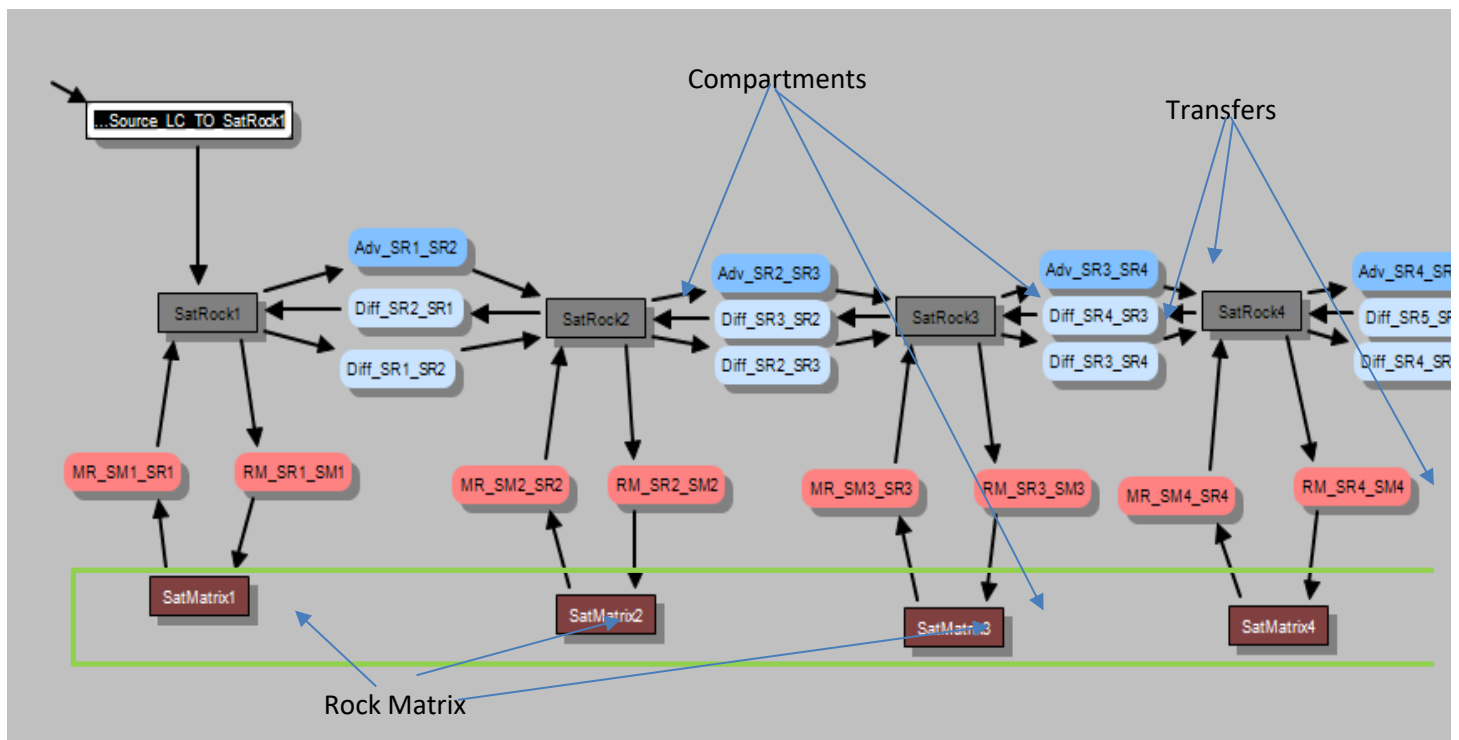

Figure 7: Discretised Geosphere showing Compartments with the transfers as implemented in AMBER

INPUT DATA FOR SIMULATION

Table 2: Radionuclides Inventory considered in the study

Radionuclide	Activity (Bq)[*]	Half Life (years)
Co-60	2.5e+14	5.27
Cs-137	2.1e+11	30
Sr-90	7.2 e+9	29.1
Am-241	8.0 e+10	432
Ra-226	10.0e+9	1600

[*]Activity modified slightly from the activity of these radionuclides in the national inventory

Table 3: Physical parameters used in the model

Hydrogeology parameters for Schist Rock		
Parameter	Value	Source
Hydraulic Conductivity _Matrix	3.65e-4m/y	After Lewis, 1989
Hydraulic Conductivity _Fracture	3.65e-2m/y	After Lewis, 1989
Hydraulic Gradient	0.05	After K. Attobra, 1983
Water-Filled Porosity _Matrix	1e-6	The Open University 2006
Water-Filled Porosity -Fracture	1e-2	The Open University 2006

RESULTS AND DISCUSSION

The assessment results for the calculated peak dose for liquid releases for a receptor dwelling around an abstraction well 5km away from the repository from the ingestion of well water from the abstraction well is presented in Fig. 8;
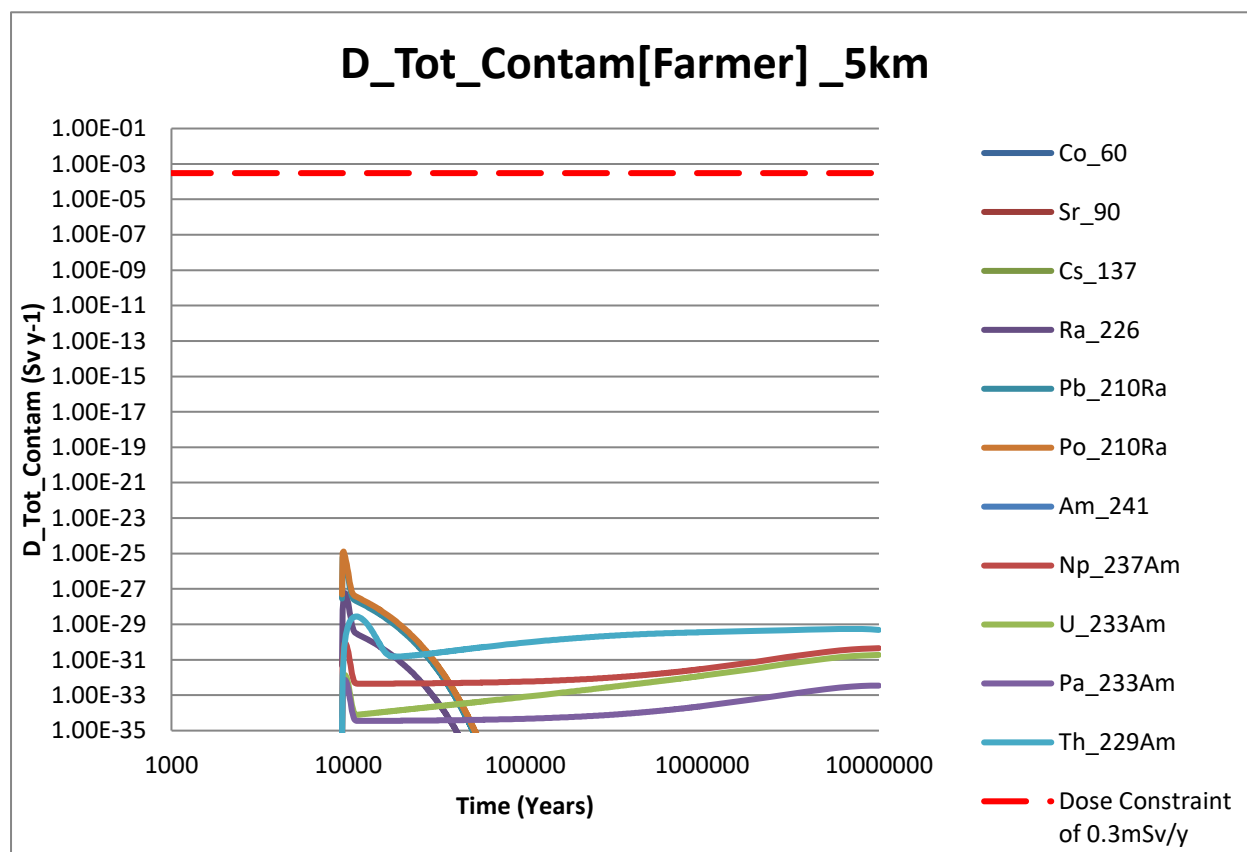

Figure 8: Calculated Dose from the Radionuclides at 5km from the Repository

From the figure, the peak dose of 2.07E-25mSv/y from long-lived Po-210, a daughter of Ra-226 occurs around 9400 years. This peak dose as compared to the dose constraint of 0.3mSv/y is insignificant.

Alternative shorter distances were considered in variant calculations. The results of which have been shown in Figs. 9a-c:
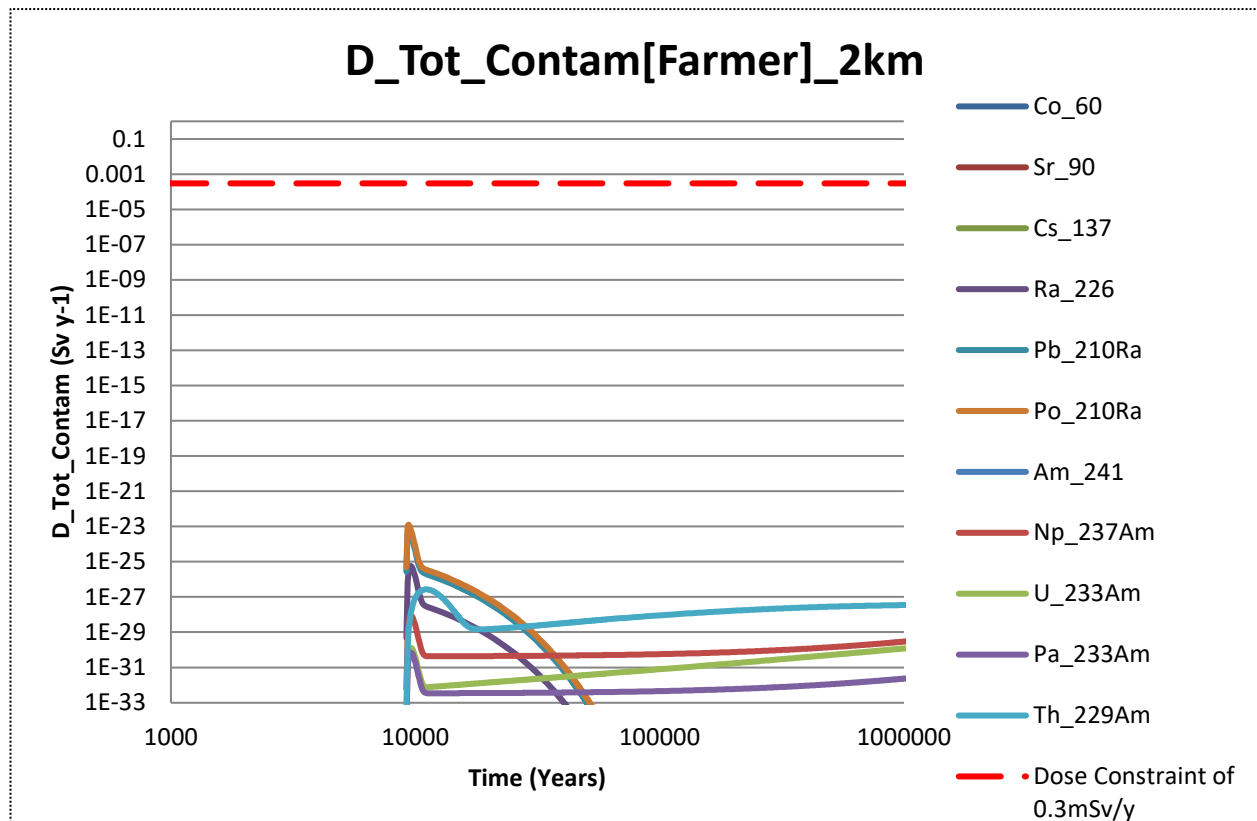
Figure 9a: Calculated Dose from the Radionuclides at 2km from the Repository
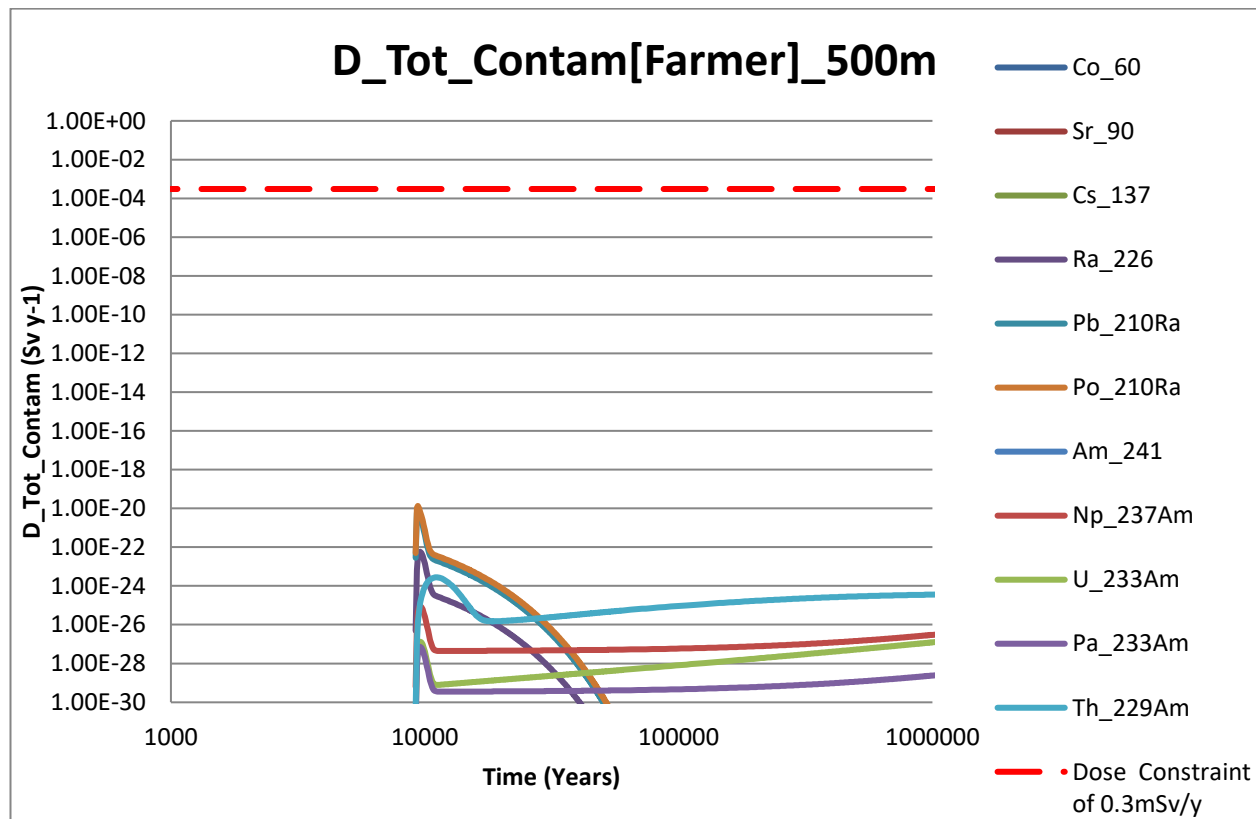

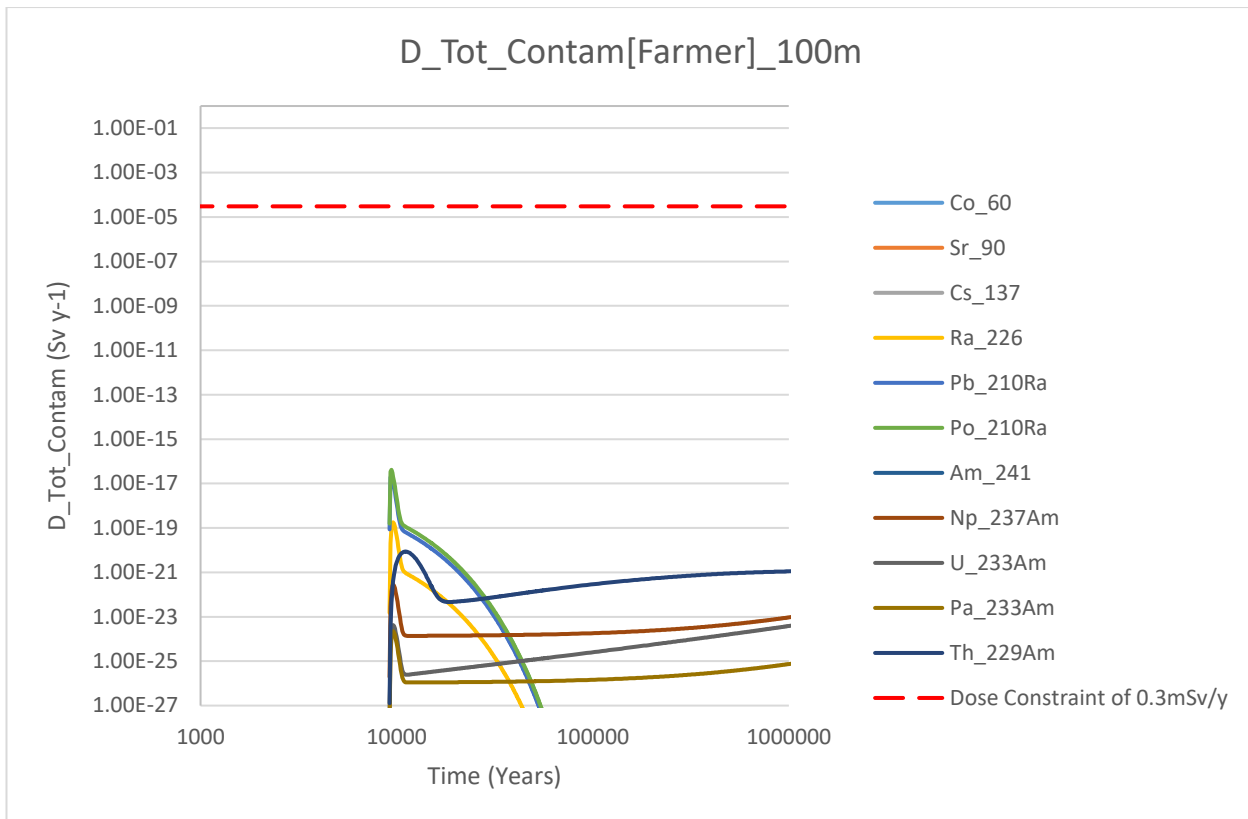Figure 9b: Calculated Dose from the Radionuclides at 500m from the Repository

Figure 9c: Calculated Dose from the Radionuclides at 100m from the Repository

From Figures 9a-c, it could be seen that the peak dose in all cases is significantly below the dose constraint of 0.3mSv/y

CONCLUSION

The quantitative results are expressed as indicative dose rates from consumption of well water as activity releases from the repository through the geosphere to the biosphere. The radioactive decay tends to reduce the concentrations of radionuclides with short half-life such as Co-60 (with a half-life of 5.27 years), Cs-137 (with a half-life of 30.0 years) and Sr-90 (with a half-life of 29.1 years) more efficiently. As can be seen from the graphs, the peak release rate comes from Po-210 a daughter from the radioactive decay of Ra-226, a long lived radionuclide with high rates of release and dose. Thus, radionuclides generated by decay are among those with higher impact on the individual by drinking water containing radionuclides released from the repository. In this simulation, Po-210 is the nuclide with greater contribution to dose, but its value is below the permissible limit of 0.3mSv/year. Based on the findings from the study, the following conclusions were drawn;

- The predicted results for liquid releases in terms of the calculated peak total dose at various receptor locations in all cases were significantly lower than the dose constraint of 0.3mSv/y.
- The results thus indicate that the Accra Plains is capable of hosting a disposal facility for Ghana's disused radioactive sources.

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