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Pollution

Elixir Pollution 41 (2011) 6041-6044

ISSN: 2229-712X

Solute transport model for radioisotopes in layered soil

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ARTICLE INFO

Article history:

Received: 21 October 2011;

Received in revised form:

17 December 2011;

Accepted: 29 December 2011;

Keywords

Geological map,

Layered soil,

Moisture propagates.

ABSTRACT

The study considered the transport of a radioactive solute in solution from the ground surface down through the soil to the groundwater when there is an accidental or intentional spillage of a radioactive material on the surface. The finite difference method was used to model the spatial and temporal profile of moisture content in a soil column using the θ-based Richard's equation leading to solution of the convective-dispersive equation for non-adsorbing solutes numerically. A matlab code has been generated to predict the transport of the radioactive contaminant, spilled on the surface of a vertically heterogeneous soil made up of two layers to determine the residence time of the solute in the unsaturated zone, the time it takes the contaminant to reach the groundwater and the amount of the solute entering the groundwater in various times and the levels of pollution in those times. The model predicted that, when there is a spillage of tritium, on the surface of the ground at the study area, it will take two years for the radionuclide to enter the groundwater and fifteen years to totally leave the unsaturated zone. Therefore, it is very important that proper containment measurements be put in place to take care of such spillages since they have long term effect.

Introduction

The particular phenomenon of concern for this work is the transport of radioisotopes in solution in the unsaturated zone of a layered soil when there is a deliberate or accidental spillage of radioactive materials on the ground.

Large quantities of radioactive materials are used in agriculture, industry, and medicine. The intended use of many of these chemicals is limited to their area of application. The accidental or intentional spillage of these radioactive chemicals on soil surfaces lead to their transfer to greater soil depths making them unavailable or ineffective for their original use, but may have an effect on the quality of nearby surface or groundwater resources.

The contamination of soil, surface and ground water has been a matter of concern worldwide. Once released into the subsurface environment, industrial and agricultural chemicals as well as radioisotopes from nuclear installations are usually subjected to a large number of immediate physical, chemical, and biological processes, including sorption-desorption, volatilization, photolysis, and biodegradation, as well as their kinetics. One special group of degradation reactions involves decay chains in which solutes are subject to sequential (or consecutive) decay reactions (Chiou, 1989).

The transport of solutes (water-soluble chemicals) through porous media is coupled with the flow of water.

Problems of solute transport involving sequential first-order decay reactions frequently occur in soil and groundwater systems. Examples are the migration of various radionuclides, the simultaneous movement of interacting nitrogen species, organic phosphate transport, and the transport of certain pesticides and their metabolites (Simdnek and van Genuchten ,1995)

The complex factors that control the movement of radioactive solute in the vadose zone and the resulting behaviour of contaminant plumes are commonly difficult to assess because of the interaction of the many factors that affect the extent and rate of contaminant movement. Predictions of movement and behaviour can be used only as estimates, and modelling is often a useful tool to integrate the various factors.

Objectives

The main objective of this study was to model the transport of a radioisotope, spilled on the surface of a vertically heterogeneous soil made up of different layers in the vicinity of the Ghana Atomic Energy Commission (GACE) reactor site at Kwabenya, Accra, to determine

- the movement of the solute in the unsaturated zone
- the time taken for the radioactive solute to reach the ground water and
- the amount of the solute entering the groundwater in various times and the levels of pollution in those times

Fig.1 Geological map of Greater Accra Region (source: CSIR-Water Research Institute, Ghana, 2009)

The study area, GAEC, Kwabenya campus falls in the Accra Plains, Ghana, (Fig.1), which is within the dry equatorial zone. The area experiences two rainfall maxima with mean annual rainfall between 74 and 89 cm; but distinct or marked dry

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seasons. The highest mean monthly temperature of about 30°C occurs between March and April and the lowest of about 26°C in August. The highest mean monthly relative humidity does not exceed 75%, and the lowest is about 60% (Dickson and Benneh, 2004).

The area is mainly underlain by acidic Dahomeyan and Togo rocks. The acidic Dahomeyan group of rocks generally consists of muscovite-biotitic gneiss, quart-feldspar gneiss, augen gneiss and minor amphibolites. These rocks weather to slightly permeable calcareous clay. The Togo rocks are mainly metamorphic and highly folded arenaceous group of rocks consisting of quartzite, quartz schist, sandstone, shale, sericite schist and phyllite. Also jasper and hematite quartz schist are common (Darko *et al*., 1995).

The area is characterised by various pedogenetic associations of clay and sandy-clay. These include the savannah ochrosols, lateritic sandy soils, Akuse series, tropical grey earths, sodium vleisols and the coastal sandy soils (Darko *et al*., 1995).

Methodology

Analysis based on resistivity-Gamma logs and vertical electrical sound studies conducted earlier on boreholes at the site gave the bedrock at a depth of 14m with two layers resulting in the physical model shown below:

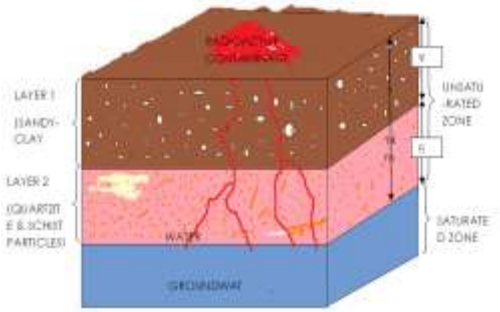

Fig. 2 Modelling Domain

As illustrated in (Fig. 2), there is spillage of radioactive contaminant on the surface of the earth. The unsaturated zone is made up of two layers, that is, layer (1) is made up of sandy clay soil and layer (2), quartzite and schist particles. The water table is at a depth 14 meters from the ground surface. There is water flow from the surface of the soil downwards towards the groundwater. The radioactive contaminant is dissolved with the water and transported towards the groundwater by convection, diffusion and dispersion and the flow is one-dimensional.

The finite difference method was used to model the spatial and temporal profile of moisture content in the soil column using Richard's equation (for vertical flow) given by (Or D. and J.M. Wraith, 2002)

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial z}\left(D.\frac{\partial \theta}{\partial z}\right) - \frac{\partial K}{\partial z} \quad (\theta - based) \qquad \ldots\ldots\ldots\ldots (1)$$

D[L^2/T] - soil water diffusivity,
K[L/T]- hydraulic conductivity,
θ[L^3/L^3] - volumetric water content,
z[L]- distance, positive downwards and
t[T] – time;
to determine the moisture content distribution in the modelling domain.

With the water content at the various depths known, the Convective-Dispersive Equation for non-adsorbing, degrading solute, given by(Or D. and J.M. Wraith, 2002):

$$\frac{\partial(\theta\, c)}{\partial t} = -\frac{\partial}{\partial z}(v\theta c) + \frac{\partial}{\partial z}(D_e\frac{\partial c}{\partial z}) - \theta\lambda c \qquad \ldots\ldots\ldots (2)$$

c[M/L^3] - solute concentration in solution;
t[T] - time;
z[L] - distance, positive downwards;
D_e[L^2/T]- effective diffusion-dispersion coefficient;
v[L/T] -mean pore-water velocity and
λ[T-1] - first order decay coefficient of the radioisotope;
was solved using finite difference method for the vertical distribution of the contaminated solute with time down to the water table .
The radioisotope considered was Tritium (^{3}H)

Results and Discussions

It is assumed that as a result of the nuclear fallouts, the surface of the soil is contaminated by 7.2g of Tritium (^{3}H) that is transported in infiltrating water deep into the soil profile. It is necessary to determine the quantity of the radionuclides that reaches the groundwater after a specified period of time. Experimental measurements in this case allow determining the concentration of the radionuclide in the soil at a certain depth and their content in the groundwater but do not allow prediction of the development of the situation in the future.

Physical-chemical soil properties determining the process of transport were set for the soil column. The influence of climatic factors was taken into account by the definition of the mean monthly temperature and rainfall for the study area.

A matlab code was developed to solve the resulting equations for the vertical distribution of the moisture content and solute concentration with time down to the water table shown in Fig. 3 and Fig. 4 respectively:

From Fig.3, the moisture content at the head of the soil column was maintained at 0.48m^3 /m^3 and that at the bottom (water table) was maintained at 0.4m^3 /m^3 with 0.01m^3 /m^3 initial moisture content for the entire modelling domain. The results of the computation show that the moisture propagates into the soil with a fairly sharp front ranging from 0.2m^3/ m^3 to 0.72m^3/ m^3.

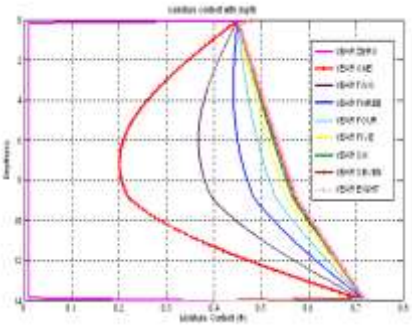

Fig. 3 Moisture Content vs. Depth

The simulation was done for sixteen years and the results are presented in Figure 4.2. It can be seen from the graph that in two years we have about 0.0011g/m3 of tritium percolating the groundwater and the value increases with years until the 8th year when it starts to reduce. This is due to the fact that, as the years go by, the radionuclide is decaying as it is being transported downwards into the groundwater.

Also the amount of the solute reaching a particular depth at a point in time can easily be obtained from the graph.

From the figure the amount of tritium entering the ground in the 4th year is small compared to the amount that entered in the 2nd year. This is based on the fact that, the quantity of tritium on the soil surface decreases as the years go by as some have

already entered the ground and some too have decayed. At the beginning of the 6th year, for instance, nothing is left on the surface of the ground.

At the beginning of the 8th year, no amount of the radionuclide is left in the zone from the surface to 2.5m depth, as the solute is moving towards the groundwater and there is nothing on the surface.

This trend continues in the ensuing years until the 16th year where no amount of the radionuclide is left in the entire modelling domain. It is clear from the figure that one year after the spillage of 7.2g of Tritium on the surface of the earth, there is no danger of the radionuclide entering the groundwater; it is after the 1st year that it starts to enter the ground waters.

Looking at the radionuclide distribution in the soil for the assessment period of 16 years, most part of the non-decayed radionuclide remain in the 1st layer; i.e. 0-9m and is available for plants uptake during root's absorption hence contaminating such plants.

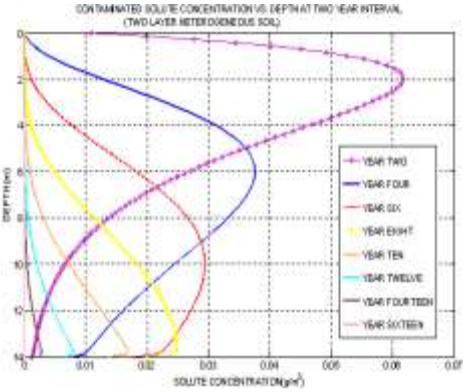

Fig. 4 Tritium Concentrations vs. Depth

Analysis of Quantity of Contaminant that Entered the Groundwater for the Entire Modelling Period

It can be seen from Fig. 5 that it will take the radionuclide two years to reach the groundwater.

Year seven and year eight recorded the maximum contamination. It is clear from the graph that one year after the spillage of 7.2g of Tritium on the surface of the ground, there is no danger of the radionuclide entering the groundwater; it is after the first year that it starts to enter the ground waters.

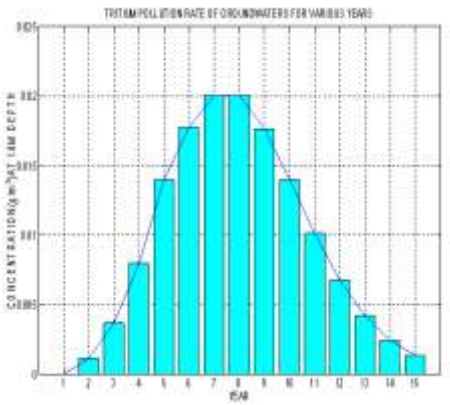

Fig. 5 Tritium Pollution Density of Groundwater for Various Years

Conclusions

The model has provided information on;

- the residence time of the radioactive solute in the unsaturated zone,
- the time taken for the radioactive solute to reach the groundwater and
- the amount of the solute entering the groundwater in various times and the levels of pollution in those times when there is a spillage of a radioactive contaminant on the surface of the ground.

From the analysis of the results, the model has been able to achieve the objectives of the study.

In particular, the resulting profiles from the simulations indicate that:

- it will take 15 years for the radionuclide to completely leave the unsaturated zone
- the amount of contamination per year into the groundwater starts after the 2nd year and
- the amount of contamination per year into the groundwater increases until the 8th year and then decreases till the 16th year. The 7th and 8th years recorded the maximum contamination.

The model predicted that the spillage of tritium on the surface of the ground poses serious long term threat to groundwater quality.

The study has also given rise to some important conclusions regarding contaminant modelling and understanding of flow and transport in the unsaturated zone. These are:

- the application of computer modelling for the analysis and prediction of the behaviour of the radioactive contaminant in the unsaturated zone and in groundwater systems can be helpful in providing relevant information on the possible level of groundwater contamination. Thus, in case there is radioactive spillage; the model can be used to estimate the quantity of solute contaminant that has reached a particular depth for the necessary action to be taken.
- with the availability of information about different soil types and long –term hydro-metrological conditions for a definite area and geographic information systems, the model can be used to monitor the vertical transport of any radioactive contaminant in the area when the need arises.

Recommendations

The following recommendations have been made for further consideration by the appropriate authorities:

- there is the need to avoid the spillage of radioactive contaminants on the ground surface as this has long term effect.
- the study revealed that much of the non-decayed contaminant remains in the first layer for a long time. There is therefore the need for work to be done to assess the quantity of the contaminant that is taken up by plants as well as the health implications on consumption of such plants.
- additional research is required to monitor the horizontal migration or transport of the contaminant in the first layer of the soil when such an accident occurs.

A major constraint in the development of the model is the validation on the basis of field observations. The limitation of field data made it difficult for the results to be adequately validated. There is therefore the need for field work to be conducted to provide good field data to thoroughly evaluate the model.

Nevertheless, the model is limited to only non adsorbing solutes and one dimensional transport (vertical flow).

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