Numerical simulation of uranium transport in flooded underground mines

Sabine M. Spiessl[1], Henning Prommer[2], Martin Sauter[1], Chunmiao Zheng[3]

[1]Institute of Geoscience, Friedrich-Schiller-University Jena, Germany.
[2]Centre for Applied Geoscience, University of Tübingen, Germany and Faculty of Civil Engineering and Geosciences, Delft University of Technology, The Netherlands.
[3]Department of Geological Science, University of Alabama, USA.

Abstract. Former mining areas generally suffer numerous environmental problems such as groundwater contamination with heavy metals and acidification. A reactive hybrid transport model has been developed to allow the quantification of uranium transport in flooded underground mines. This hybrid model specifically takes into account two different transport time scales within an underground mine, which result from the existence of a network of highly conductive shafts, drifts or ventilation raises within the considerably less permeable ore material in the underground. This paper introduces the model concept and addresses potential model applications.

Introduction

The occurrence of acidic, metal-rich groundwater is often observed in mining areas. At sites of uranium mining, exposure of receptors to radioactivity might result from the migration of radionuclides within the subsurface. In many cases the oxidation of naturally abundant pyrite has shown to be the key process that causes the acidic geochemical environment (Wunderly et al. 1996). Thus, in the last few years, underground or subsurface mines have undergone controlled flooding to prevent access of oxygen and thereby the further lowering of pH due to pyrite oxidation (Bain et al. 2001). Subsurface mines are typically composed of networks of highly conductive shafts, drifts or ventilation raises within a considerably less permeable ore material. Therefore, the simulation of contaminant transport in such domains requires consideration of two distinct time scales in the transport model. As a result of the discrete nature of a pipe network, and because the quantification of contaminant flux from the solid mineral phase into the mobile water phase requires a discrete surface area, double continuum models cannot be employed for modelling of reactive transport in underground mines. Therefore, a “hybrid“ transport model has been developed and subsequently coupled with a geochemical model. The underlying concepts of the resulting numerical model RUMT3D (i.e., three-dimensional reactive underground mine transport model) are the focus of the present paper as well as a demonstration of its potential use.

Hybrid flow model (CAVE)

To compute the flow field in the combined pipe-network/matrix system, the CAVE model (Clemens et al. 1996), a hybrid model coupling flow in a discrete conduit network to a continuum is employed. CAVE was originally developed for modelling the genesis of karst aquifers. However, the simulation of groundwater flow in underground mines, i.e. a system with both, a highly conductive conduit network and a considerably less permeable, porous matrix can be treated in an analogous way as karstified carbonate aquifers.

The three-dimensional continuum equation for flow in the ore material (matrix), including a further source/sink term γ that couples the continuum to the pipe-flow model, is described as follows:

$$\frac{\partial}{\partial x_m}\left(K_{m,xx}\frac{\partial h_m}{\partial x_m}\right)+\frac{\partial}{\partial y_m}\left(K_{m,yy}\frac{\partial h_m}{\partial y_m}\right)+\frac{\partial}{\partial z_m}\left(K_{m,zz}\frac{\partial h_m}{\partial z_m}\right)-W_m-\gamma=S\frac{\partial h_m}{\partial t_m} \qquad (1)$$

with x_m, y_m, z_m [L] the distance along the respective Cartesian coordinate axis in the ore material, t_m [T] represents the time, h_m [L] is the hydraulic head in the ore material, $K_{m,xx}$, $K_{m,yy}$, $K_{m,zz}$ [L T^{-1}] depict the values of hydraulic conductivity along the coordinate axes in the ore material, S [L^{-1}] is the specific storage coefficient, W_m [T^{-1}] is a volumetric flux term per unit volume from a sink/source into the ore material e.g. groundwater recharge, and γ [T^{-1}] denotes the volumetric rate of fluid transfer between the ore material and the conduit system per unit volume.

A conduit system is defined in the model as a pipe network consisting of cylindrical tubes. Conduit nodes are introduced between the connecting tubes to allow for exchange of flow between the different tubes from different faces of a cell and between a conduit node and the continuum (matrix, ore material) at different locations in the model domain. Only one conduit node can be placed in an ore material cell. There are 6 potential faces of a cell so of a conduit node for tube connection in 3-dimensional model domains, i.e., top, bottom, front, back, left and right. A conduit tube may extend over one or more ore material (matrix) cells depending on the respective geometry of the mineshafts or adits, and the locations of the sinks and sources, e.g. direct recharge and fixed heads. Conduit orientations can be freely designed, i.e. they do not necessarily need to be vertical or horizontal along the same continuum layer, so that the model can easily match the actual spatial coordinates of real mine networks. It is assumed that the conduit system is fully saturated, an assumption that will hold for most flooded underground mines. Flow between the ore material and the conduit nodes is described by a linear relationship between the two systems (Barenblatt et al. 1960):

$$\Gamma_i = \alpha_i \left(h_i - h_{i,m} \right) \tag{2}$$

where Γ_i [L^3 T^{-1}] represents the exchange flow rate between conduit node *i* and the ore material cell, α_i [L^2 T^{-1}] stands for the exchange coefficient between node *i* and the ore material, h_i [L] is the hydraulic head at conduit node *i* and $h_{i,m}$ [L]is the hydraulic head in the ore material cell where conduit node *i* is located. The magnitude of exchange coefficient α_i depends on the hydraulic conductivity of the ore material and geometrical factors, determined by the discretization of the adjacent continuum cell.

Laminar flow in each tube, i.e., laminar flow from one conduit node to another can be calculated from an expression for discharge which can be found by substituting Hagen-Poiseuille's into Darcy-Weisbach equation. Turbulent flow is solved according to the implicit Colebrook-White law. Conservation of flow volume at any conduit node *i* can be determined by using Kirchhoff's law, which states that the total inflow and outflow must balance at each node. A more detailed description of the pipeflow model can be found in the work by Clemens et al. (1996).

Hybrid transport model (UMT3D)

Transport of mass within the ore material (matrix) is also simulated with a continuum approach, i.e. with the standardized three-dimensional multi-species transport model MT3DMS (Zheng and Wang 1999). In contrast, mass transport within the network of tubes is modelled discretely with a one-dimensional advective transport model. Coupling between the continuum transport model and the transport in the conduits is achieved using a sequential operator-splitting procedure (Walter et al. 1994, Steefel and MacQuarrie 1996). The transport of solutes in the continuum is solved first, followed by a second step during which the solute concentrations in the conduit system are updated and the solute mass is transported. The coupled hybrid transport model is UMT3D (three-dimensional underground mine transport model).

The general transport equation (Zheng and Wang 1999) has been extended to include a further sink/source term, i.e. ξ to couple MT3DMS to the conduit transport model. Thus, the three-dimensional transport equation in the continuum is:

$$\frac{\partial(\theta_m C_m^k)}{\partial t_m} = \frac{\partial}{\partial x_{m,i}} \left(\theta_m D_{m,ij} \frac{\partial C_m^k}{\partial x_{m,j}} \right) - \frac{\partial}{\partial x_{m,i}} \left(\theta_m v_{m,i} C_m^k \right) + q_{m,s} C_{m,s}^k + \xi^k + \sum_{k=1}^{N} RXN_{m,k} \tag{3}$$

with C_m^k [M L^{-3}] is the aqueous concentration of component *k* in the ore material, $D_{m,ij}$ [L^2 T^{-1}] depicts the hydrodynamic dispersion coefficient tensor in the ore material, $v_{m,i}$ [L T^{-1}] is the linear pore water velocity in the ore material, $q_{m,s}$ [T^{-1}] denotes the volumetric flux of water per unit volume of ore material representing sources (positive) and sinks (negative), θ_m [-] is the porosity of the ore material, $C_{m,s}^k$ [M L^{-3}] is the concentration of component *k* of the sources or sinks to the ore material, ξ^k [M L^{-3} T^{-1}] denotes the volumetric rate of fluid transfer between the ore material and the conduit system per unit volume multiplied by the respective concentration values of the conduit sink and source terms of component *k* and $RXN_{m,k}$ [M L^{-3} T^{-1}] is the chemical reaction term of the ore material with respect to component *k*.

For the calculation of solute transport from the continuum cell to a conduit node *i*, the mass transfer rate is determined by multiplying the cell concentration of the pore water in the ore material of component *k*, $C_{m,i}^k$, at node *i*, with the respective exchange flow rate Γ_i and dividing the mass flux by the volume of the respective cell, $V_{m,i}$, i.e.,

$$\frac{\Gamma_i C_{m,i}^k}{V_{m,i}} \tag{4}$$

Alternatively, if solute mass is transported from conduit node *i* into the matrix cell, then $C_{i,m}^k$ is replaced by the nodal concentration, C_i^k. Fig.1 illustrates this exchange for both cases. Such mass transfer rates are then treated as mass sink/source terms in MT3DMS since conduit nodes act similarly to other sinks/sources (e.g., wells) in MT3DMS. The difference between wells as implemented in MT3DMS and conduit nodes is that the mass removed by means of a well is not returned to the ore material, while the mass removed with an entry conduit node may be returned completely through the exit conduit nodes. Amount, location and required time for these returns mainly depend on (i) the transport velocity in the different conduits, (ii) the magnitude of the exchange coefficients between the exit conduit nodes and the ore material, (iii) the magnitude of conduit sink terms, and (iv) the length of the different conduit tubes.

A one-dimensional transport equation that solely considers advection is applied to each tube, i.e.,

$$\frac{\partial C_j^k}{\partial t} = -q_j \frac{\partial C_j^k}{\partial z_j} \tag{5}$$

where C_j^k [M L^{-3}] represents the aqueous concentration of component *k* in conduit tube *j*, q_j [L T^{-1}] is the flux of water in tube *j*, *t* [T] is the time and z_j [L] refers to the distance along a respective Cartesian coordinate axis in the respective tube *j*. Note, there are no sink/source terms in this transport equation. The mass exchange rates from the ore material, from the six potentially connecting tubes and from conduit sink/source terms such as direct recharge, fixed head and fixed concentration as applied to the different conduit nodes are considered in terms of initial or boundary concentration values to the transport equation (5). The resulting concentration values are obtained by a weighted arithmetic mean of the single flow and transport components for each transport time step. Such an approach is common in mixing cell models (Bajracharya and Barry 1993). Mathematically, a weighted arithmetic mean of the concentration value of component *k* at conduit node *i* can be expressed as follows:

$$C_i^k = \frac{\sum_f^6 Q_{i,j}^{f+} C_{i,j,l}^{k,f} + \Gamma_i^+ C_{i,m}^k + \sum_s Q_{i,s}^+ C_{i,s}^k}{\sum_f^6 Q_{i,j}^{f+} + \Gamma_i^+ + \sum_s Q_{i,s}^+} \tag{6}$$

where $Q_{i,j}^{f+}$ [L^3 T^{-1}] represents the discharge of tube *j* connected to face *f* of conduit node *i* into the respective node *i,* $Q_{i,s}^+$ [L^3 T^{-1}] refers to a volumetric flow rate of a conduit source term to node *i* , and the subscript *l* refers to the first or the maximum number of tube sections or segments in the different tubes depending on the flow direction. The tubes or conduits can further be divided into a user-defined number of segments or sections to decrease the effect of numerical dispersion and improve numerical stability. Also note that tubes within a network can be considerably longer than cell widths, lengths or thickness.

Eq. (5) is solved with a mass-conservative semi-Lagrangian scheme (EMCNOT). This scheme was developed by Liu et al. (2001) for modelling advection-dominated mass transport problems and is an explicit mass conservative scheme without time step limitation. With the EMCNOT method, a transport time step size of up to the minimum residence time value of a respective pipe in the conduit system multiplied by a user defined Courant number, *Cr* can theoretically be used for both models. The residence time of groundwater in a specific pipe transported by advection under steady state flow conditions is determined by dividing the length of a pipe, L_j by the flow velocity in a respective pipe. In mathematical form, the maximal transport time step size, Δt_{max} can be expressed as:

$$\Delta t_{\max} = Cr \left(\frac{L_j}{q_j} \right)_{\min} \tag{7}$$

Such a time criterion may vary with each flow time step since the flow rate in each tube may change with each flow time step. Spiessl et al. (2002) demonstrated that applying the EMCNOT scheme to solve advective transport in the conduit system significantly reduces numerical dispersion compared to the standard finite difference (FD) method.

Mass balance in the conduit system is determined in a similar way as in the ore material. To check performance of both transport models, mass balance calculations are also carried out independently for the different conduit nodes in MT3DMS as sink/source terms. In contrast to the conduit transport model by Birk (2001) the above-described model uses a global approach to calculate mass balance and thus improves computational efficiency. Moreover, the global variable arrays and subroutines in the modified conduit transport model are fully compatible with those used in MT3DMS.

Reactive hybrid transport model (RUMT3D)

The comprehensive geochemical model PHREEQC-2 (Parkhurst and Appelo 1999) was coupled with the UMT3D model as a solver for the reaction term within Eq. (3), i.e., $RXN_{m,k}$ using a sequential operator splitting technique approach (Walter et al. 1994, Steefel and MacQuarrie 1996). The transport equation (3) does not have to be solved for every individual chemical species, but only for total aqueous component concentrations (Yeh and Tripathi 1989, Engesgaard and Kipp 1992), defined as:

$$C_u = c_u + \sum_{k=1,n_s} Y_k^s s_k \tag{8}$$

where C_u is the total aqueous component concentration of the u^{th} component, c_u is the molar concentration of the u^{th} (uncomplexed) aqueous component, n_s is the number of dissolved species that form complexes with the u^{th} aqueous component, Y_k^s is the stoichiometric coefficient of the aqueous component in the k^{th} complexed species and s_k is the molar concentration of the k^{th} complexed species. The (local) redox-state, *p*e, is at present modeled by transporting chemicals/components in different redox states separately, while the *p*H is calculated based on a (local) charge balance. The transport model UMT3D needs to solve transport for n_{tot} entities, with

$$n_{tot} = n_{e,nre} + \sum_{k=1,n_{e,re}} n_{rs,k} \tag{9}$$

where $n_{e,nre}$ is the number of (mobile) chemical elements occurring in only one redox state, $n_{e,re}$ is the number of elements occurring in multiple redox states and $n_{rs,k}$ is the appropriate number of different possible redox states of the k^{th} element. The resulting model (RUMT3D) can handle a wide range of chemically reactive processes including aqueous complexation, mineral dissolution/precipitation, and ion-exchange. Reactions might be assumed to occur as equilibrium reactions and/or kinetically controlled. More details on the incorporation of PHREEQC-2 and MT3DMS can be found in Prommer et al. (2002). Chemical reactions in the conduit system are at present assumed to have a negligible effect on the composition of the groundwater because of the typical short residence times of the solutes in the conduit system.

Scenarios of reactive transport in a coupled conduit-continuum system

In order to demonstrate the capabilities of the RUMT3D model two simplified, schematic scenarios, one of which includes the hydraulic effect of a single U-shaped conduit system have been selected. Solute fluxes (discharge of uranium species) to a river in the proximity of a subsurface mine were compared for a model setup (i) natural groundwater flow conditions and (ii) flow conditions affected by the presence of a pipe network. For both scenarios, steady state groundwater flow and reactive transport in a vertical cross-section was simulated with a continuum (matrix, ore material) domain of 1200 m by 480 m. Heads at the river were fixed at 390 m, imposing a hydraulic gradient between ore material and the discharge point. A fixed constant-head boundary of 470 m was set at the “influent” boundary of the model. A uniform groundwater recharge rate of 6x10^{-9} m s^{-1} was assumed at the surface. The ore matrix has a vertical and a horizontal hydraulic conductivity of 1.0x10^{-6} m s^{-1} and an effective porosity of 0.05.

Table 1. Chemical composition of the background groundwater, the uranium/pyrite ore body, the influent boundary and the recharge water.

Component	Background groundwater, uranium/pyrite ore body and influent boundary (mol/l)	Recharge water (mol/l)	Component	Background groundwater, uranium/pyrite ore body and influent boundary (mol/l)	Recharge water (mol/l)
pH	7.53[a]/7.53[b]/7.24[c]	7.66	S(6)	8.010×10^{-3}	1.000×10^{-3}
pe	-3.20[a]/-3.07[b]/-3.11[c]	12.99	S(-2)	1.474×10^{-12}	-
C(4)	1.220×10^{-3}	1.447×10^{-3}	U(3)	-	-
Ca	5.225×10^{-3}	1.947×10^{-3}	U(4)	-	-
Cl	1.100×10^{-3}	1.000×10^{-3}	U(5)	-	-
Fe(2)	1.001×10^{-3}	6.030×10^{-16}	U(6)	-	-
Fe(3)	5.010×10^{-10}	1.000×10^{-5}	Calcite	1.000[a,b]	-
Na	5.788×10^{-3}	5.000×10^{-4}	Pyrite	(1.000×10^{-3})[b]	-
O(0)	-	7.000×10^{-4}	UO_2(am)	(1.000×10^{-3})[b]	-

[a] for background groundwater only

[b] for uranium/pyrite ore body only

[c] for influent boundary only.

A uranium-pyrite rich ore body was placed in the centre of the flow domain as depicted in Fig. 2. The remaining continuum cells are assumed not to contain any amorphous uranium oxide or pyrite. An anaerobic homogeneous water composition was assumed at the beginning of the simulation. The reaction network consists of 15 aqueous components (see Table 1) and three minerals (amorphous uranium oxide, pyrite, calcite). In contrast to uranium oxide and pyrite, calcite was assumed to be uniformly distributed within the aquifer. Table 1 also lists the chemical composition of the background groundwater, the uranium oxide/pyrite ore body, of the influent water at the constant-head boundary and of the recharge water.

In one of the two simulations a conduit system was placed into the model domain just below the water table (see Fig.2) to investigate its influence on the uranium mobilization process. The U-shaped conduit system consists of two vertical shafts and one horizontal drift modelled with 49 conduit nodes and 48 tubes. The tubes have a diameter of 0.5 m. The horizontal drift was located in the centre of the uranium/pyrite ore body where mining activities are likely. The flow simulations show that approximately half of the conduit nodes in the pipe network are entry nodes (all on the left side of the conduit system) while the other half are exit nodes. Figs.3a and 3b show the head profiles of the flow set-up without and with the conduit system, respectively. The equipotential 450 m in Fig.3b separates entry from exit nodes. For the (assumed) exchange coefficient of 1.0x10^{-5} m^2 s^{-1}, flow conditions remain laminar throughout the conduit system.

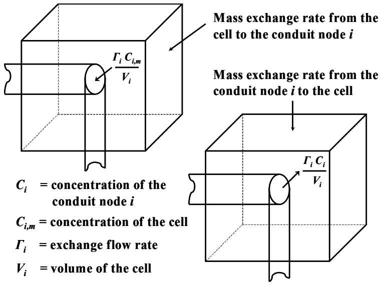

Fig. 1. Illustration of ore material cell-conduit exchange.

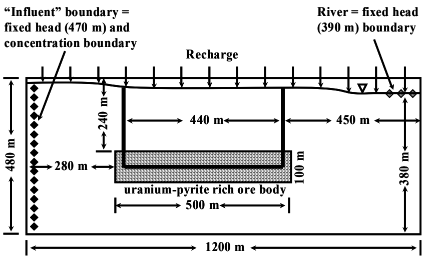

Fig. 2. Configuration of the simplified, schematic mine system.

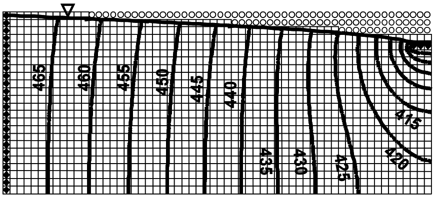

Fig. 3a. Head distribution profile for the mine system without conduit system.

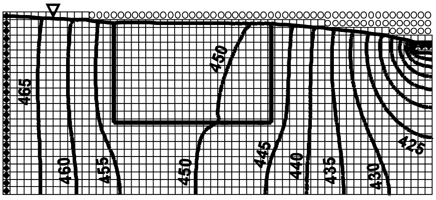

Fig. 3b. Head distribution profile for the mine system with conduits.

Initial results

Preliminary simulation results of RUMT3D indicate that the conduit system plays a major role for the release and the transport of uranium species to the discharge point. As apparent from Fig.4b, U(VI) (e.g., UO_2^{2+}) is mobilised in the area surrounding the conduit system and rapidly discharged into the river. With no conduit system present the U(VI) species remain stable within the ore body (Fig.4a), i.e. only negligible concentration/mass of uranium can be observed downgradient from the ore body.

Fig. 4a. U(VI) concentration profile for the mine system without conduit system.

Fig. 4b. U(VI) concentration profile for the mine system with conduits.

Summary and future work

This paper presents the reactive hybrid transport model RUMT3D developed for three-dimensional reactive underground mine transport problems. RUMT3D can handle two different transport time scales, found in underground mine systems. It is planned to further integrate surface controlled release of uranium into the model by implementing kinetically controlled reaction rate equations into the PHREEQC-2 database. Furthermore, sorption to charged surfaces (surface complexation) e.g. to iron(III) oxides will certainly play an important role for the mobility of uranium and will thus need consideration in future work. The assumption of a negligible effect of chemical reactions in the conduit system due to much shorter residence times of the components in the conduit system than in the ore material may also require further investigation.

Acknowledgements

The funding for this study was mainly provided by the European Commission (PIRAMID project, Contract EVK1-CT-1999-000021). Thanks are also given to Gaisheng Liu who prepared the numerical method EMCNOT to use for the solution of advection in the conduit transport model.

References

Bajracharya K., Barry D.A. (1993). Mixing cell models for nonlinear equilibrium single species adsorption and transport. J. Contam Hydrology 12: 227-243.

Barenblatt G.I., Zheltov I.P., Kochina I.N. (1960). Basic concepts in the theory of seepage of homogeneous liquids in fissured rocks. J Appl Math and Mech 24: 1286-1303.

Bain J.G., Mayer K.U., Blowes D.W., Frind E.O., Molson J.W.H., Kahnt R., Jenk, U. (2001). Modelling the closure-related geochemical evolution of groundwater at a former uranium mine. J Contam Hydrology 52: 109-135.

Birk S. (2001): Characterisation of karst systems by simulating aquifer genesis and spring responses: model development and application to gypsum karst. Geowissenschaftliche Arbeiten, C60, Tübingen, Germany.

Clemens T., Hückinghaus D., Sauter M., Liedl R., Teutsch G. (1996). A combined continuum and discrete network reactive transport model for the simulation of karst development. Proc ModelCARE 1996, Golden, Colorado, IAHS Publ. 237: 309-318.

Engesgaard P., Kipp K.L. (1992). A geochemical transport model for redox-controlled movement of mineral fronts in groundwater flow systems: A case of nitrate removal by oxidation of pyrite. Water Resour Res 28: 2829-2843.

Liu G., Wang P., Zheng C. (2001). An explicit and mass-conservative scheme without time-step limit for modeling advection-dominated contaminant transport, 2001 MODFLOW international conference in Golden, Colorado.

Parkhurst D.L., Appelo C.A.J. (1999). User's guide to PHREEQC (version 2) – a computer program for speciation, batch-reaction, one-dimensional transport, and inverse geochemical calculations. Water-Resources Investigations Report 99-4259, U.S. Dept. of the Interior, U.S. Geological Survey.

Prommer H., Bajyrry D.A., Zheng C. (2002). MODFLOW/MT3DMS based reactive multicomponent transport modelling. Accepted for publication in Ground Water.

Spiessl S.M., Sauter M., Zheng C., Liu G. (2002). Simulation of contaminant transport in flooded underground mines using a coupled continuum-conduit transport model: Comparison of two numerical methods for advection in the pipe network. Submitted to Proc ModelCARE 2002, Prag, 17-20 June 2002.

Steefel C.I., MacQuarrie K.T.B. (1996). Approaches to modeling of reactive transport in porous media. In: Reactive Transport in Porous Media (ed. by P. C. Lichtner P.C., Steefel C.I., Oelkers E.H.) Min. Soc. Of America, 1996, 83-129.

Walter A.L., Frind E.O., Blowes D.W., Ptacek C.J., Molson J.W. (1994). Modeling of multicomponent reactive transport in groundwater. 1. Model development and evaluation. Water Resour Res 30: 3137-3148.

Wunderly M.D., Blowes D.W., Frind E.O., Ptacek C.J. (1996). Sulfide mineral oxidation and subsequent reactive transport of oxidation products in mine tailings impoundments: A numerical model. Water Resour Res 32: 3173-3187.

Yeh G.T., Tripathi V.S. (1989). A critical evaluation of recent developments in hydrogeochemical transport models of reactive multichemical components, Water Resour Res 25: 93-108.

Zheng C., Wang P.P. (1999). MT3DMS: A modular three-dimensional multispecies model for simulation of advection, dispersion and chemical reactions of contaminants in groundwater systems; Documentation and User's Guide, Contract Report SERDP-99-1, U.S. Army Engineer Research and Development Center, Vicksburg, MS.