Subsurface Multiphase Flow and Multicomponent Reactive Transport Modeling using High-Performance Computing

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Abstract. Numerical modeling is a critical tool to the U.S. Department of Energy for evaluating the environmental impact of remediation strategies for subsurface legacy waste sites. Unfortunately, the physical and chemical complexity of many sites overwhelms the capabilities of even most state of the art groundwater models. Of particular concern is the representation of highly-heterogeneous stratified rock/soil layers in the subsurface and the biological and geochemical interactions of chemical species within multiple fluid phases. There is clearly a need for higher-resolution modeling (i.e. increased spatial and temporal resolution) and increasingly mechanistic descriptions of subsurface physicochemical processes (i.e. increased chemical degrees of freedom).

We present SciDAC-funded research being performed in furthering the development of PFLOTRAN, a parallel multiphase flow and multicomponent reactive transport model. Written in Fortran90, PFLOTRAN is founded upon PETSc data structures and solvers. We are employing PFLOTRAN to simulate uranium transport at the Hanford 300 Area, a contaminated site of major concern to the Department of Energy, the State of Washington, and other government agencies. By leveraging the billions of degrees of freedom available through high-performance computation using tens of thousands of processors, we can better characterize the release of uranium into groundwater and its subsequent transport to the Columbia River, and thereby better understand and evaluate the effectiveness of various proposed remediation strategies.

1. Introduction
Over the past several decades, subsurface (groundwater) flow and transport models have become vital tools for the U.S. Department of Energy (DOE) in its environmental stewardship mission. These models have been employed to evaluate the impact of alternative energy sources and the efficacy of proposed remediation strategies for legacy waste sites. For years, groundwater models remained arguably simple in comparison to the sophisticated models of today. These traditional models simulated single-phase groundwater flow and single-component solute transport within a single continuum, with basic chemical reactions such as radioactive decay and linear sorption to rock/soil surfaces. Although these simplified groundwater models are still widely used today, advances in subsurface science have enabled the development of more sophisticated models that
employ multiple fluid phases and chemical components coupled through a suite of biological and geochemical reactions at multiple scales. However, with this increased complexity comes the need for more computational power, typically beyond that of the average desktop computer. This is especially true when applying these sophisticated multiphase flow and multicomponent reactive transport models to two- or three-dimensional problem domains.

The advent of high-performance computation has not only satisfied this computational need, but also provided the computational resources necessary to further increase the complexity and sophistication of today’s subsurface models. Although supercomputers provide the raw computing power and storage facilities necessary to execute large and complex simulations in the subsurface, few groundwater models actually leverage high-performance computation on a regular basis. Some may argue that the limited availability of computing platforms on which to execute large-scale simulations is to blame, but the cost of implementing efficient parallel algorithms is most likely the root of the problem. Since groundwater model developers often lack the skills necessary to parallelize their models, they have to outsource the parallelization effort to computational scientists. For those who attempt parallelization without the assistance (and cost) of parallel programming expertise, their codes rarely scale well when running on hundreds to thousands of processors. Thus, the quandary at hand is how to develop and maintain a parallel subsurface groundwater model in a collaboratory effort that unites subsurface and computational scientists at a minimal cost.

In this paper, the multiphase flow and multicomponent reactive transport code PFLOTRAN is presented as a successful implementation of high-performance computation within an existing subsurface groundwater model, and a brief history is provided regarding circumstances that enabled this accomplishment. As part of the DOE SciDAC program, PFLOTRAN is undergoing continued development and support to demonstrate the utility of high-performance computation in groundwater modeling. As part of SciDAC, PFLOTRAN is being employed to better answer high-priority subsurface science questions for DOE. These include (1) modeling the viability of subsurface sequestration of carbon dioxide as a potential means of alleviating global warming due to greenhouse gas emissions, and (2) simulation of uranium transport within the Hanford 300 Area in southeastern Washington State to quantify the physical and chemical processes that impact the leaching of uranium from subsurface sediments and subsequent flow into the Columbia River. This paper presents the latter scenario at the Hanford 300 Area where a large, preliminary tera-scale simulation of the site was executed on a supercomputer.

2. Background on Groundwater Modeling Theory

Although a wide variety of formulations exist for multiphase fluid flow in porous media, the simplified governing equation for variably-saturated groundwater flow based on Richards equation is presented here:

$$\frac{\partial}{\partial t} (\phi s \rho) + \nabla \cdot \rho \mathbf{u} = S.$$  \hspace{1cm} (1)

The fluid flux $\mathbf{u}$ in Equation 1 is computed as a function of Darcy’s law

$$\mathbf{u} = -\frac{\kappa \kappa_r}{\mu} \nabla (p - pgz),$$ \hspace{1cm} (2)

with pressure $p$, vertical distance $z$, porosity $\phi$, saturation $s$, permeability $\kappa$, relative permeability $\kappa_r$, fluid viscosity $\mu$, fluid density $\rho$, acceleration of gravity $g$, and a source/sink term $S$. Since the relative permeability $\kappa_r$ is a nonlinear function of saturation $s$, Equation 1 is nonlinear and the resulting nonlinear system of equations must be solved using an iteration scheme such as Picard iteration, or more preferably Newton’s method.

The governing equation for multicomponent solute transport has the form

$$\frac{\partial}{\partial t} (\phi \psi_i) - \nabla \cdot (\phi D \nabla \psi_i - \rho \psi_i \mathbf{u}) = \mathcal{R}_i(e_1, e_2, \ldots, e_N) + S_i,$$ \hspace{1cm} (3)
where the total component concentration $\psi_i$ of species $i$ is a nonlinear function of the $N_c$ primary chemical component concentrations $c_i$ in the system provided through thermodynamic mass action relations. Other quantities appearing in this equation represent the Darcy flux $u$, diffusion/dispersion coefficient $D$, chemical reaction rates $R_i$ (functions of primary species concentrations $c_i$), and a source/sink term $S_i$. When chemical components are coupled through nonlinear reaction terms, the resulting system of equations is also nonlinear and must be solved with a nonlinear solution scheme such as Newton Raphson.

Although the multiphase flow and multicomponent reactive transport equations can be solved simultaneously, most models utilize a sequentially coupled approach where flow is first solved for the pressure head and subsequent Darcy fluxes, and then chemical components transported and reacted. For the coupling of transport and chemical reaction, three primary approaches are utilized. These include (1) the global implicit approach where transport and reaction are solved in a single, tightly coupled system of equations, (2) the sequential iteration approach where transport and reaction are solved iteratively, until the loosely-coupled system converges to within prescribed tolerances, and (3) the sequential non-iterative approach (more commonly referred to as operator splitting) where each chemical component is transported separately and then chemical reactions are solved based on the chemical component concentrations within each grid cell/node. The sequential non-iterative approach is perhaps the most common coupling approach among large, high-performance codes since it does not require the solution of large, coupled systems of equations spanning the entire problem domain, while the global implicit approach is predominantly limited to one-dimensional models, though the computing resources available through high-performance computation have enabled the use of the global implicit approach on large and complex reactive transport problems (e.g. \[1\]).

In short, when simulating subsurface flow and transport, the modeler develops a conceptual model of the subsurface domain and discretizes it across a computational grid or mesh using the governing equations presented above. He or she then assigns parameters to the domain (e.g. permeability, chemical reaction coefficients, etc.) along with boundary and initial conditions (e.g. pressure, temperature, and aqueous and mineral concentrations) based on available field and laboratory data. The model then takes discrete steps through time solving the resulting systems of equations and storing simulation results at select locations in space and time. In the end, analysis of the simulation results may be used to inform decision makers, enabling better predictions of groundwater flow and transport under different management schemes.

3. PFLOTRAN

PFLOTRAN is a parallel multiphase flow and multicomponent reactive (geochemical) transport code that originated from the serial FLOTRAN code developed at Los Alamos National Laboratory \[2\]. The code is composed of flow and transport modules (PFLOW and PTRAN, respectively), with the option of running the modules in coupled or decoupled mode. PFLOW is capable of simulating fluid flow through porous media with the following fluid phases: air, water, supercritical CO$_2$. PFLOW-generated fluid flow velocities or fluxes are utilized by PTRAN to compute solute transport. Within PTRAN, transport and reaction are fully coupled. PFLOTRAN's problem domain is discretized spatially using the integrated finite volume approach, with fully-implicit backward-Euler time differencing.

The development of PFLOTRAN speaks to the success of the Computational Science Graduate Fellowship (CSGF) Program administered by the Krell Institute for the U.S. Department of Energy. As part of their summer practicum assignments, fellows Glenn Hammond (1999-2003) and Richard Mills (2001-2004) worked with Lichtner to incorporate the SciDAC TOPS-funded PETSc library \[3\] within FLOTRAN's flow and transport modules, resulting in a parallel version of the code, or PFLOTRAN \[1, 4\]. (Essentially, the PFLOW and PTRAN modules were each parallelized over a summer and have been continually enhanced since.)
multiphase modules within PFLOW were developed later by Lichtner and Chuan Lu [5, 6].

PFLOTRAN’s parallel paradigm is based on domain decomposition where the computational problem domain is divided into subdomains, one domain assigned to each processor employed. PFLOTRAN then leverages PETSc solvers and data structures to link these subdomains within a single parallel code. Since PFLOTRAN is founded upon these PETSc data structures, the code is capable of utilizing the full suite of algorithms available in the library. In addition, PETSc also provides linkage to a variety of external software packages (e.g. Hypre, Trilinos, Zoltan). Within PFLOTRAN, the distributed array (DA) is the key PETSc data structure that dictates the layout and decomposition of the three-dimensional parallel domain. From the DA, PETSc generates parallel matrices (Mat) and vectors (Vec) with consistently mapped local and global indexing for the parallel communication required to solve the problem. Therefore, although an indepth understanding of parallel communication paradigms such as the message passing interface (MPI) is helpful, it is not required since PETSc hides the communication from the end user. For example, when employing the Newton-Raphson method to solve a nonlinear system of equations in parallel using the PETSc nonlinear solver or SNES, the programmer essentially provides functions to PETSc that evaluate the residual and compute the Jacobian for the nodes that reside locally on each processor. PETSc then utilizes these functions to compute a solution vector, which is then returned to PFLOTRAN for boundary condition updates, I/O, etc. Thus, the application scientist is able to focus more on the science (in this case, subsurface physics and chemistry) rather than solvers, preconditioners, etc.

4. Application of PFLOTRAN
4.1. Hanford 300 Area: Background
As part of the SciDAC groundwater science application area, PFLOTRAN is being utilized to simulate radionuclide transport at the U.S. Department of Energy’s Hanford Site in southeastern Washington State. Throughout the Cold War, Hanford served as a facility within the DOE nuclear weapons complex for the production of plutonium for national defense. Plutonium production initiated during the latter years of World War II and continued until the 1980s, after which efforts at the Hanford Site shifted toward environmental remediation.

Of particular interest to DOE is the cleanup/remediation of the Hanford 300 Area, which is located in the southeast corner of the Hanford Site, immediately north of the city of Richland along the Columbia River. During the Cold War, facilities at the Hanford 300 Area were utilized to research and fabricate nuclear fuels for plutonium production at the Hanford Site. As a result of this fuel production, liquid and solid wastes were often disposed of in process ponds and process trenches within the confines of the Hanford 300 Area. This waste is known to have contained uranium, though the exact amount released is uncertain. Over time, this waste has migrated downward to the water table polluting the underlying groundwater.

Significant simulation and modeling efforts were put forth in the early 1990s in an attempt to determine the rate at which uranium within the groundwater would migrate toward and discharge into the Columbia River [7]. Unfortunately, these models lacked the sophistication necessary to accurately simulate uranium transport, predicting that the uranium would be removed or flushed out of the Hanford 300 Area subsurface by ambient groundwater flow within a decade. Today, nearly 15 years later, uranium concentrations at the site remain relatively unchanged [8]. Several theories exist attempting to explain the discrepancies between the predicted and observed transport of uranium. These include (1) inadequacies in the uranium transport conceptual model due to overly simplistic reactions representing the sorption of uranium on sediment grains and (2) uncertainty of the uranium source term mass concentrations and leaching rates. Since these theories involve uncertainties in both the flow and reactive transport calculations employed, improvements in the accuracy of both process models must be found.
The conceptual model for flow at the Hanford 300 Area is complicated by the highly permeable Hanford Unit and the rapidly fluctuating Columbia River producing changes in magnitude and direction of flow. The soils beneath the Hanford 300 Area are composed of layers of high and low permeability soil, the top layer of which is the highly permeable Hanford Unit located near the water table. The Hanford Unit is composed of highly permeable cobbles, gravels, and sands. Hydraulic conductivities within the Hanford Unit are on the order of a thousand to tens of thousands of meters per day. In comparison, the Ringold Units below the Hanford Unit exhibit much lower hydraulic conductivities of 0.01 to 150 meters per day. Thus, within the Hanford Unit, where all original uranium sources are believed to have resided, groundwater has the potential of flowing rapidly with very small pressure gradients in the aquifer.

Rapid fluctuations in river stage, as shown in Figure 1, can produce these pressure gradients. Diurnal (daily) fluctuation in river stage can be up to 1.5 meters, while seasonal fluctuation can exceed 2.5 meters. The variable release of water from Priest Rapids Dam upriver is the cause of the diurnal fluctuation, while the seasonal variation is due to snow melt, irrigation, etc. This fluctuation in river stage along with the highly permeable Hanford Unit directly impacts the water table beneath the Hanford 300 Area, causing river waters to flow into the area during high stage and retreat during low stage. One could view this phenomenon as a slow tidal effect where sources of uranium residing immediately above the water table in the vadose zone are cyclicly flushed over time as the water table rises and falls, introducing new uranium mass into the underlying aquifer. An accurate depiction of the flow processes beneath the Hanford 300 Area is critical for capturing such behavior in the uranium transport model.

![Columbia River stage versus time during 1992.](image)

Figure 1. Columbia River stage versus time during 1992.

Regarding the transport of uranium, it is believed that the linear ($K_d$) approach to sorption utilized in the transport models from the early 1990s is overly simplistic, and that a more rigorous depiction of uranium sorption, such as that provided by a sophisticated nonlinear surface complexation model, may be necessary. In addition, the numerical representation of the subsurface domain may need further refinement, either through grid refinement, adaptive gridding, or sub-continua. Traditionally, hydrologic parameters are averaged over a representative elementary volume (REV) yielding a single continuum model formulation. The
REV is often the size of a model grid block. The division of the single REV into multiple coupled sub-continua has shown promise. Lichtner [9, 10] demonstrated that multi-continuum modeling provides a better fit to 1D uranium column experiments based on Hanford 300 Area sediments carried out by Oafoku et al. [11]. Thus, a more refined, mechanistic representation of the physical and chemical process scales in the Hanford 300 Area subsurface may be needed to predict the migration of the uranium plume due to transport in the groundwater.

The modeling of uranium transport at the Hanford 300 Area is a difficult challenge that will require large computational resources. To date, models depicting the site are either two-dimensional or overly coarse in 3D [12]. Parallel computation on high-performance supercomputing platforms provides the computational resources necessary to refine the Hanford 300 Area model both spatially and geochemically.

4.2. Hanford 300 Area: Modeling

As a first step in building the Hanford 300 Area numerical model, a three-dimensional model of the site was developed composed of over 2 million grid cells (135 × 250 × 60 cells). This rectangular “box” model of the site includes regions above the site’s ground surface and within the Columbia River. Aquifer material properties (e.g. permeability, porosity, variably saturated flow parameters) were mapped to the 10 × 10 × 1 meter grid cells based on each cell’s physical location in the domain and the corresponding rock/soil material type at that location. The conceptual model shown in Figure 2 illustrates the layering of the various rock/soil types below the Hanford 300 Area. To better visualize the extent of the site, an aerial view of the Hanford 300 Area is superimposed on the domain. Notice the green Hanford Unit immediately below the ground surface and underlying Ringold Units (i.e. blue, orange, cyan, yellow). These stratigraphic layers were generated from geologic data (i.e. well picks) [13].

The gridded domain spanned 1350 by 2500 meters horizontally (east-west by north-south) with the vertical dimension of the model domain spanning 60 meters, between 70 and 130 meters.

![Figure 2. Conceptual model of Hanford 300 Area hydrostratigraphic units.](image-url)
above sea level. Steady-state hydrostatic boundary conditions were set on the east and west boundaries of the model corresponding to hypothetically prescribed inland (106.5 meters, west) and river (104.5 meters, east) piezometric heads, resulting in a hydraulic gradient of $1.48 \times 10^{-3}$ (2.0 meter drop) toward the river. Zero flow boundary conditions were assigned to the remaining four boundaries (i.e. north, south, top, and bottom). Thus, steady boundary conditions were assumed with no surface recharge due to rainfall.

An initial water table was prescribed as a linearly graded plane between the east and west boundaries. To determine the potential for transport toward the Columbia River, two hypothetical plumes were initially introduced within the discretized domain near the water table interface beneath the north and south process ponds as shown in Figure 3. Each of these plumes spanned block regions in the subsurface measuring 50 meters horizontal (both east-west and north-south) and 16 meters vertical near the water table. Overall, 6,075,000 degrees of freedom (unknowns) were solved in this problem, involving 3 degrees of freedom (pressure, temperature, solute concentration) per grid cell, with 2,025,000 cells. Flow and transport was simulated for 2 years using 512 Cray XT4 processors on Oak Ridge National Laboratory’s (ORNL) Jaguar supercomputer.

![Figure 3. Initial locations of hypothetical plumes in Hanford 300 Area north and south process ponds.](image)

Simulation results demonstrated that within the first year of simulation time both plumes exited the domain across the east boundary into the Columbia River demonstrating the potential for rapid transport to the river. This result was expected since a linear gradient was prescribed across the domain between the west and east boundary conditions. It should be noted that this hypothetical flow and transport scenario does not consider the fluctuating river stage mentioned previously. The key outcome of this preliminary modeling of the Hanford 300 Area was less the accurate prediction of uranium transport and more the demonstration of a large tera-scale conceptual model executed on a supercomputing platform.

With fluctuating river stage, the hydraulic gradient across the site will most likely be much lower and will change direction with time. Additional simulations with transient boundary conditions are necessary to characterize this phenomenon, and are planned for the near future. To date, there is much uncertainty regarding the actual rate at which uranium enters the river.
However, as the sophistication of the Hanford 300 Area model increases through the addition of transient inland and river boundary conditions, surface recharge (rainfall), and stochastic representations of subsurface properties along with verification against existing modeling efforts and validation against observed data, the predictive capability of the model will improve.

4.3. Hanford 300 Area: PFLOTRAN Performance
As previously mentioned, the coupled flow and transport simulations were executed on ORNL’s Jaguar supercomputer utilizing 512 of the Cray’s XT4 processors. To evaluate the performance of PFLOTRAN on this preliminary Hanford 300 Area model, the scenario was run out to 200 simulation hours on 16, 32, 64, 128, 256 and 512 cores utilizing either 1 or 2 cores per processor with no I/O; a 4-hour restriction on processing time on \( \leq 128 \) cores limited the minimum number of cores that could be used. Figure 4 illustrates the performance of PFLOTRAN on Jaguar for the various processor configurations. From the results shown in the figures, it is clear that PFLOTRAN scales well up to 256 cores, after which performance begins to tail off with an efficiency of 65-68% at 512 cores depending on the number of cores employed per processor. Since the number of unknowns per processor approaches \( \sim 12,000 \) for the 512 processor decomposition, this tailing off is likely due to inadequate problem size.

![Figure 4. Wall clock and relative speedup curves for the Hanford 300 Area model executed on ORNL’s Jaguar XT4 nodes.](image)

The performance results shown in Figure 4 also demonstrate that limiting the number of cores per processor to 1 improves the performance of PFLOTRAN by \( \sim 10-20\% \), based on wall clock time. This slight degradation in per core performance is most likely due to memory bandwidth contention when running fully-packed processors. Obviously, the degradation in performance is only minor and all cores should be employed to maximize the utilization of available processing power since the single core per processor simulations do not utilize the operations available through the second core.

5. Future directions
To date, only preliminary simulations of the Hanford 300 Area using PFLOTRAN have been executed. The flow solution has considered only constant or steady hydrostatic boundary conditions with no recharge, while transport has been limited to a conservative tracer originating from hypothetical sources. To provide more realistic predictive simulations of the Hanford 300...
Area, enhancements to the site conceptual model need to be developed and incorporated into the PFLOTRAN model.

The current model assumes a “box” model of the site where the entire domain, including regions above the site’s ground surface and within the Columbia River, is composed of porous media. In addition, the current model assumes steady boundary conditions with no surface recharge due to rainfall. Therefore, important enhancements to the flow conceptual model include (1) the addition of transient hydrostatic boundary conditions based on river stage and inland observation well data, (2) the addition of a transient recharge boundary condition at the ground surface, and (3) inactivation of grid cells located beyond the domain of interest (e.g. above the ground surface and within the river where no porous media exists).

Major enhancements to the Hanford 300 Area transport model are necessary since the current model assumes transport of a conservative tracer emanating from hypothetical sources. Potential enhancements include: (1) the addition of uranium geochemistry composed of all pertinent uranium species in the aqueous, sorbed, and mineral phases; (2) a multicontinuum transport formulation to better capture uranium mass transfer between advection and diffusion dominated portions of the domain; and (3) a more accurate representation of the uranium source term based on a multiscale formulation using available observation data. With these enhancements to the site conceptual model, the PFLOTRAN model of the Hanford 300 Area should become increasingly accurate in its predictive capability.

6. Conclusion
As today’s subsurface flow and transport models become increasingly sophisticated, simulating an increased number of physical and chemical phenomena at higher resolutions and/or finer scales, high performance computation provides the computing power necessary to keep the solution of these models attainable. The subsurface multiphase flow and multicomponent reactive transport code PFLOTRAN, is an excellent example of the harnessing of high-performance computation through the instrumentation of an efficient parallel library (i.e. PETSc) within a legacy model. In this work, PFLOTRAN was employed to simulate variably-saturated groundwater flow and conservative tracer transport of two hypothetical plumes within the Hanford 300 Area, exhibiting excellent scalability on up to 256 processors (relative to a 16 processor wall clock time) on a problem with approximately 6 million unknowns. Future SciDAC-funded modeling at the Hanford 300 Area will entail increasing the number of physical and chemical processes being modeled (e.g. adding multiple continua, uranium geochemistry) and incorporating these enhancements within the PFLOTRAN model of the site.

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