Objectives and Tentative Scientific Approach for Uranium Reactive Transport Modeling in the 300 Area

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Conceptual Model Development and Reactive Transport Modeling for the 300 Area Uranium Plume in 300-FF-5
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Presentation Outline

I. Conceptual model underlying the existing 300-FF-5 Record of Decision

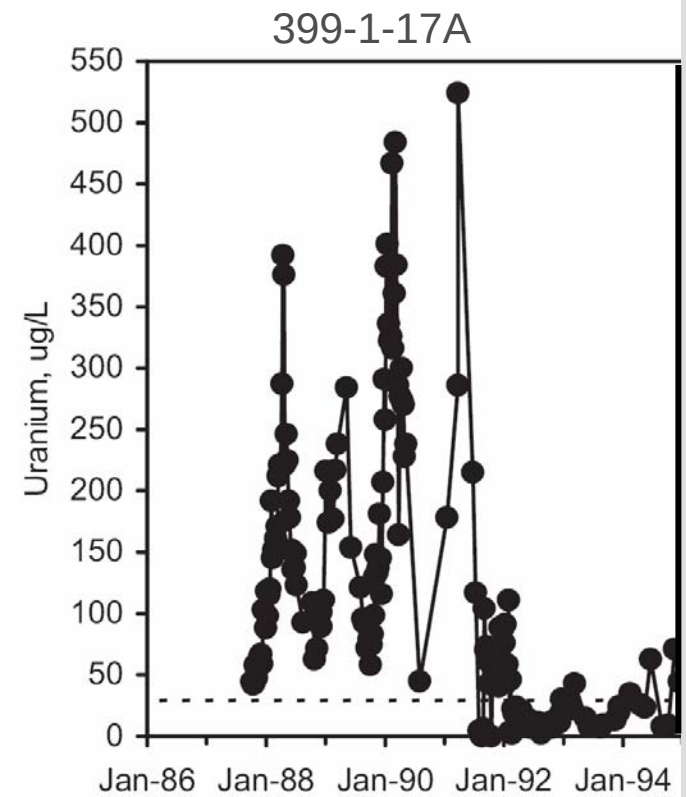
II. Assessment and update of the conceptual model

III. Proposed modeling approach

Background

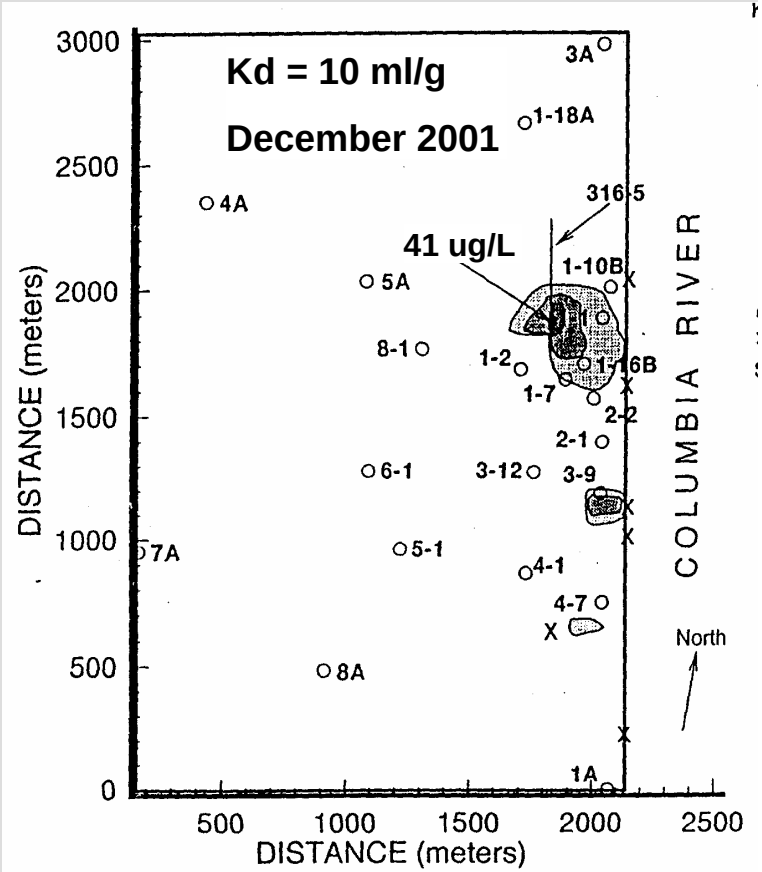
1991 Expedited Response Action

- Remove contaminated soils from process trenches
- End discharge of uranium to process trenches



1993 WHC Flow and Uranium Transport Simulation

- Predicted maximum total uranium concentrations
  - 19 ug/L in 2000 (K_d=1 ml/g)
  - 41 ug/L in 2001 (K_d=10 ml/g)



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1996 CERCLA 300-FF-5 Record of Decision

- Contaminated groundwater operable unit
- Goal: Restore aquifer to drinking water standard
- Interim Remedy: Monitored natural attenuation
  - Continued monitoring while source remedial actions ongoing
  - 5-year review of ROD in 2001: reauthorized interim remedy as remediation activities still ongoing

Motivation for Updating the 300 Area Uranium Transport Conceptual Model

- 2005 Focus Feasibility Study
- 2006 review of CERCLA 300-FF-5 ROD and final remedy
- Opportunity to assess predictions from1993 modeling study
- Several perturbations in the last 15 years
  - Removal of contaminated soils from process trenches (ERA 1991)
  - Uranium loading ends (1991)
  - Data from expanded monitoring program (1992)
  - Liquid discharges end (1994)
  - High river stage in 1997 (1996, 2002)
  - Backfilling of open trenches (2004)
- New knowledge from ongoing efforts directed at the 300 Area
  - Monitoring programs
    - Groundwater
    - Near-Shore
    - River
  - Characterization studies
    - Uranium Geochemistry
    - Geology
    - Hydrogeology
  - Modeling studies
    - DOE
    - NRC

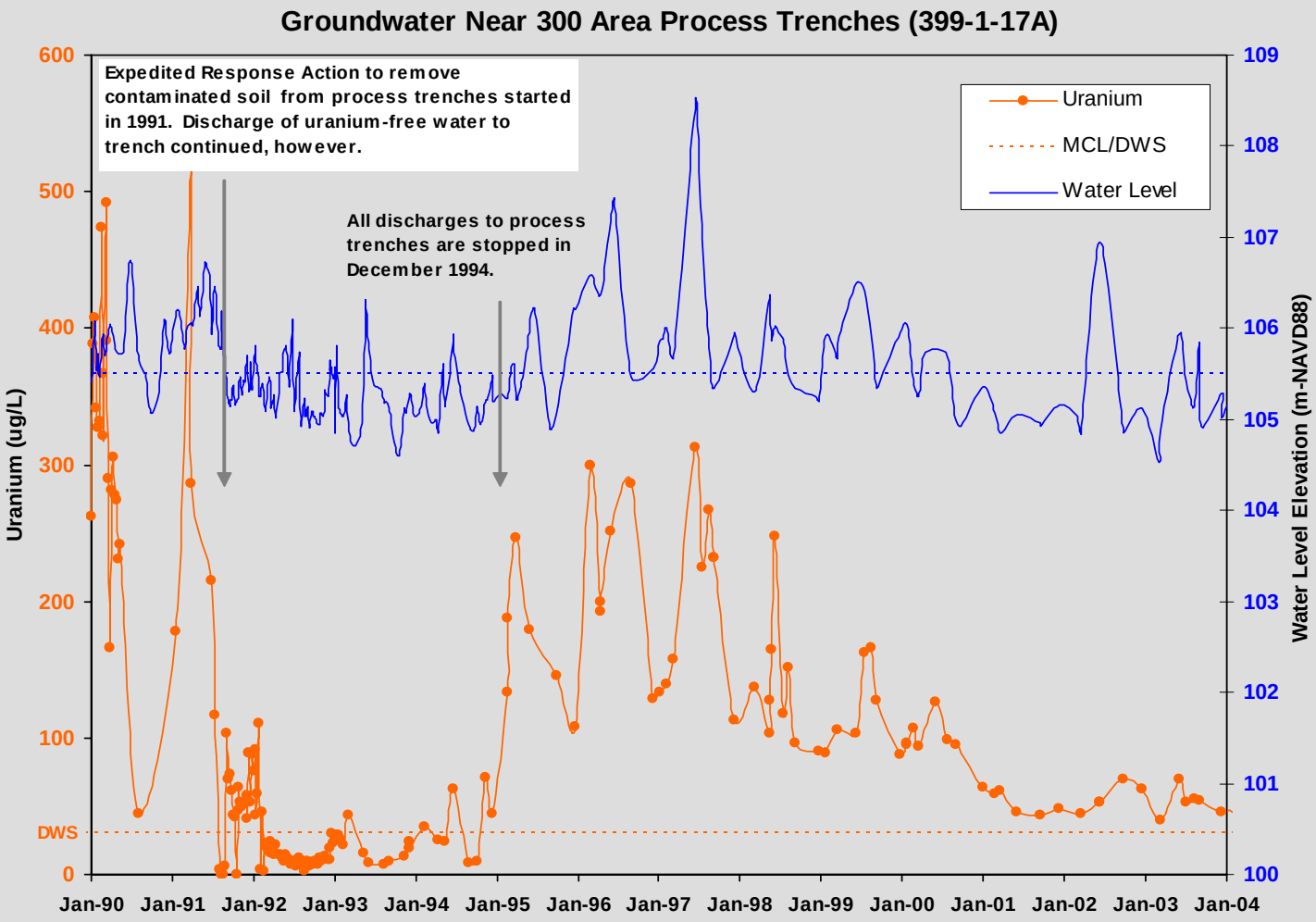
1993 WHC Modeling Approach

- Modeling Objective: Predictions of "reasonably conservative maximum values" for uranium in the groundwater between 1992 and 2018
- Spatial distribution of 4 hydrofacies types based on isopach maps (Gaylord et al. 1991)

	Hydraulic Conductivity (m/d)	Bulk Density (g/cc)	Effective Porosity (-)
Gravel	7500	2.0	0.2
Sandy Gravel	1500	1.8	0.252
Sand	2	1.4	0.382
Mud / Silt	0.2	1.2	0.462

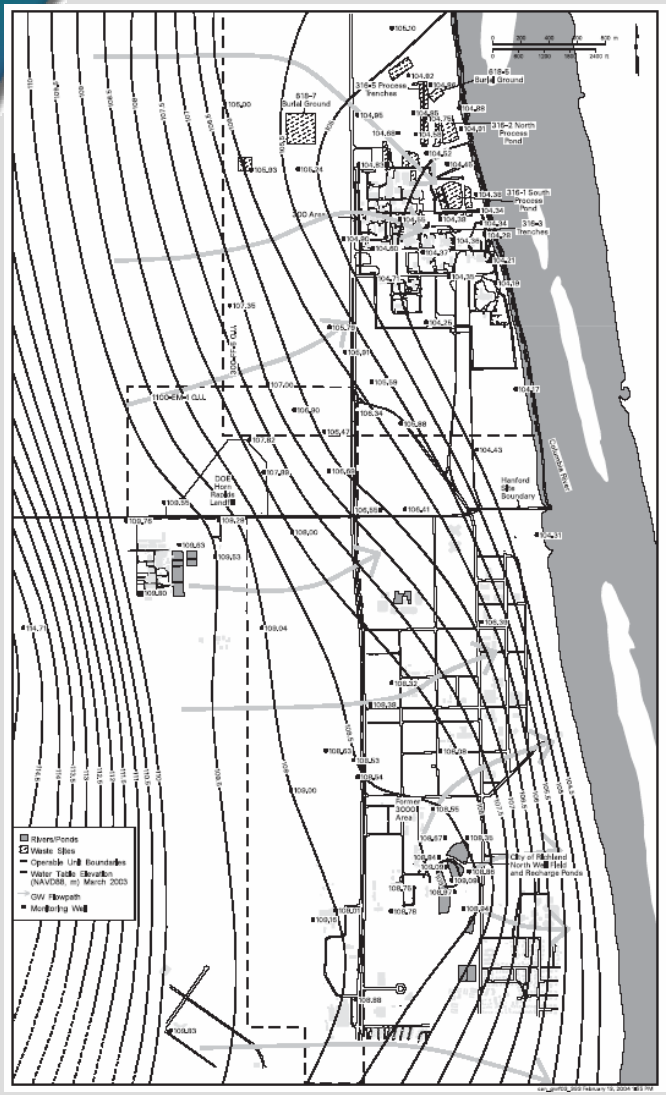
  - 3D variable spacing cartesian grid: 61 x 90 x 25 for 1978m x 3030m x 31.5m
- Assumed that source control measures would prevent surface or vadose zone contamination from impacting underlying groundwater
  - 3D flow in saturated unconfined aquifer
    - vadose zone was not modeled
    - driven primarily by monthly river stage changes
    - recharge through ground surface not modeled
    - bank storage and springs not modeled
  - transport of existing uranium plume
    - based on 1991 sampling program
    - no other uranium sources
- Uranium mobility controlled by constant K_d
  - reversible equilibrium
  - independent of water chemistry
  - independent of mineralogy

Aquifer Water Levels and Uranium Concentrations

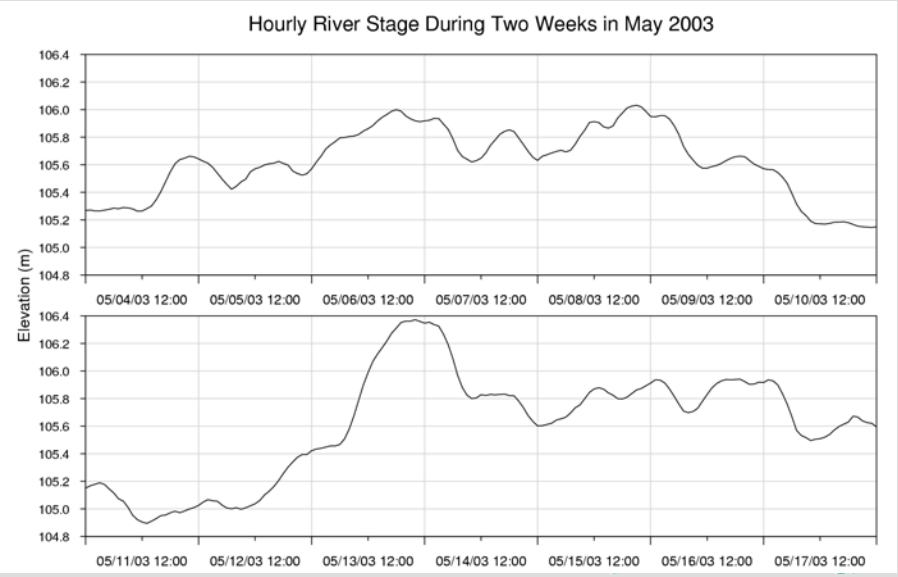


U-plume-mvnt.xls (05/04/04)

River Stage Fluctuation and Groundwater Flow



- 300 Area is in the McNary Dam backwater
  - river stage is a function of discharge at Priest Rapids and pool level at McNary
  - 0.2 to 0.6 m variation from backwater effect
- Temporal variability driven by river stage fluctuations
  - Long-term Average
    - regional: convergence of flow reflects transmissive materials in unconfined aquifer
    - local: preferential flow paths due to small scale heterogeneities within larger scale hydrofacies
  - Seasonal
    - low stage: towards river
    - high stage: stronger southerly component parallel to river
  - Hourly/Daily/Weekly
    - short-term gradient changes caused by river fluctuations
    - principal impact on mass transfer across aquifer-river interface



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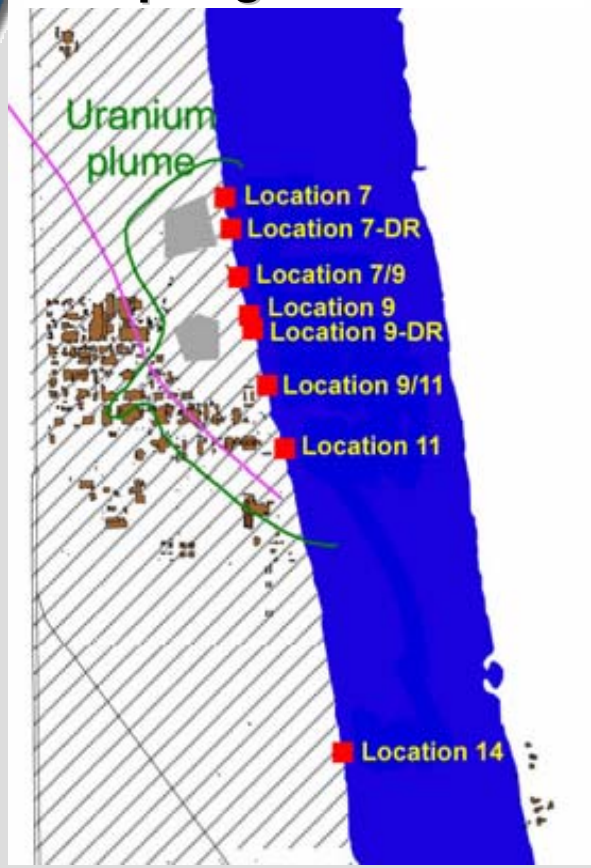
Uranium Transport from Aquifer to River (Patton et al. 2003)

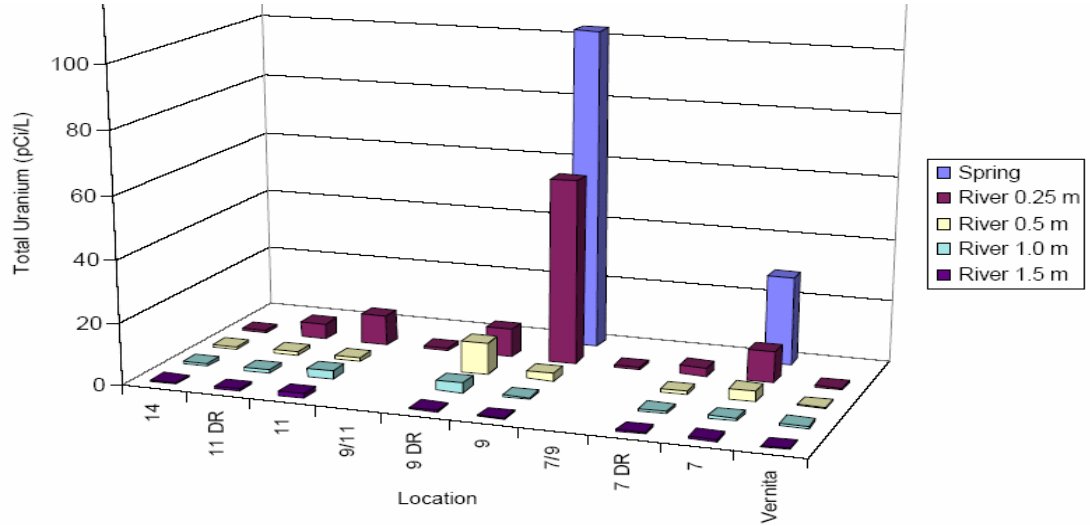
300 Area Near-Shore Sampling Locations



Riverbank Springs and Near-Shore River

- Aug – Oct 2001 (Low Stage) Near-Shore Survey
  - Focused on 4 known springs within uranium plume
  - Sampling of springs, river water
- A few riverbank springs provide preferential pathways for uranium to the near-shore 300 Area
- River water uranium (6 cm from bottom) highest near shore but always lower than spring concentrations
- In the absence of flowing riverbank springs, diffuse bank seepage can influence the concentration of uranium in river water nearest the shoreline

300 Area and Vernita Bridge Near-Shore River Water and Riverbank Spring Samples



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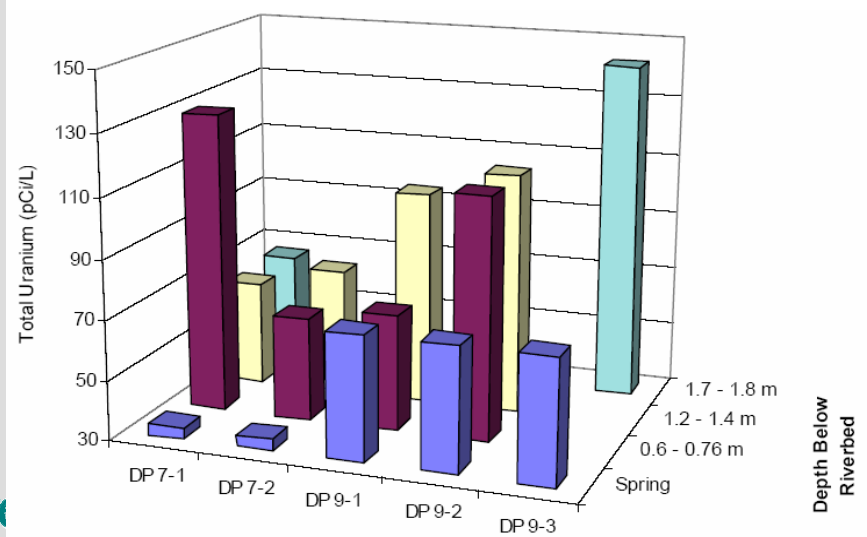
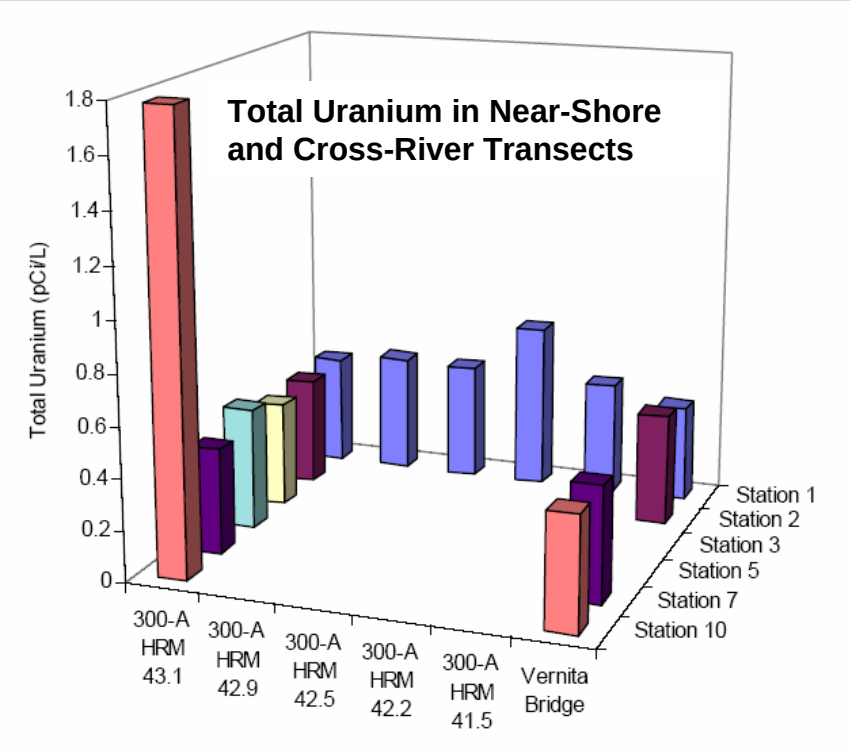
Uranium Transport from Aquifer to River (cont'd)

Near-shore riverbed (drive point) water samples

- Aug – Oct 2001 (Low Stage) Focus on spring locations
  - Location 7: two distances (4.6, 9.4 m) out into river from spring and three depths (0.6/0.76, 1.2/1.3, 1.8 m)
  - Location 9: three distances (1, 8, 14.3 m) out into river from spring at three depths (0.6, 1.2/1.4, 1.7 m) Water samples from river bed
- Upwelling of uranium associated with shallow, near-shore river water by flowing and non-flowing springs
  - All riverbed uranium concentrations were above ambient water quality criteria with values ranging from 64 to 143 pCi/L
  - no clear trend in concentration versus depth below the riverbed for the uranium values.

Total Uranium for Water Samples Collected from 300 Area Shallow Groundwater (drive point samples)





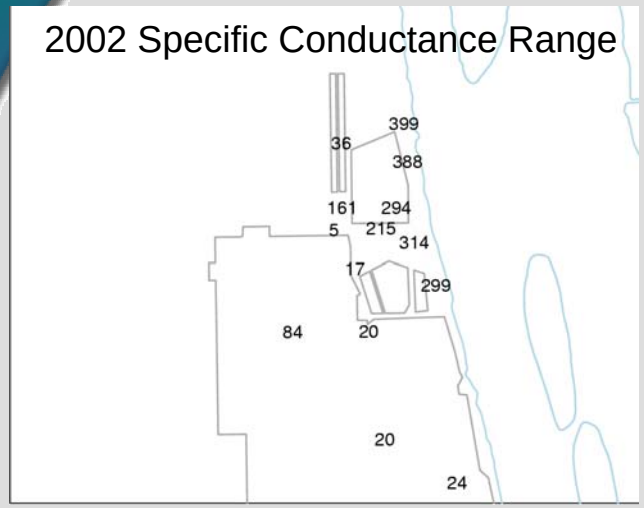
Cross-river water column transects

- Highest uranium is for the Franklin County shoreline directly across from the Richland pumphouse and is most likely related to irrigation water returns (Poston et al. 2001)

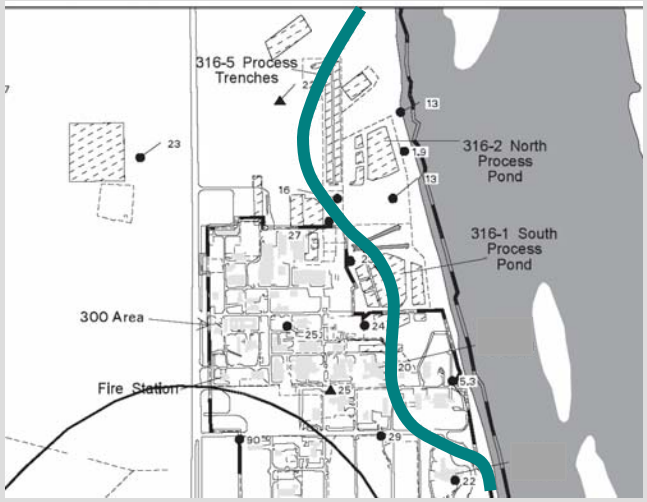
Subsurface Flow

- Key Issues:
  - Liquid migration through uranium-contaminated vadose zone sediments
    - Fundamental to characterization of continuous uranium source term
  - Liquid flux between the aquifer and river
    - Fundamental to net uranium mass flux out of aquifer
    - Relative contribution of springs/preferential flow paths versus diffuse exchange through aquifer – river interface
- Saturated-unsaturated flow model required to address
  - Spatially and temporally variable surface recharge
  - Unsaturated flow in vadose zone
  - Groundwater flow in unconfined aquifer
  - Imbibition and drainage of sediments due to fluctuating water table
  - Flow between aquifer and river
    - Influx of river water through aquifer – river interface during high stage
    - Upwelling of aquifer water through aquifer – river interface during low stage
  - Riverbank storage and dynamic seepage face formation due to hourly fluctuations in river stage

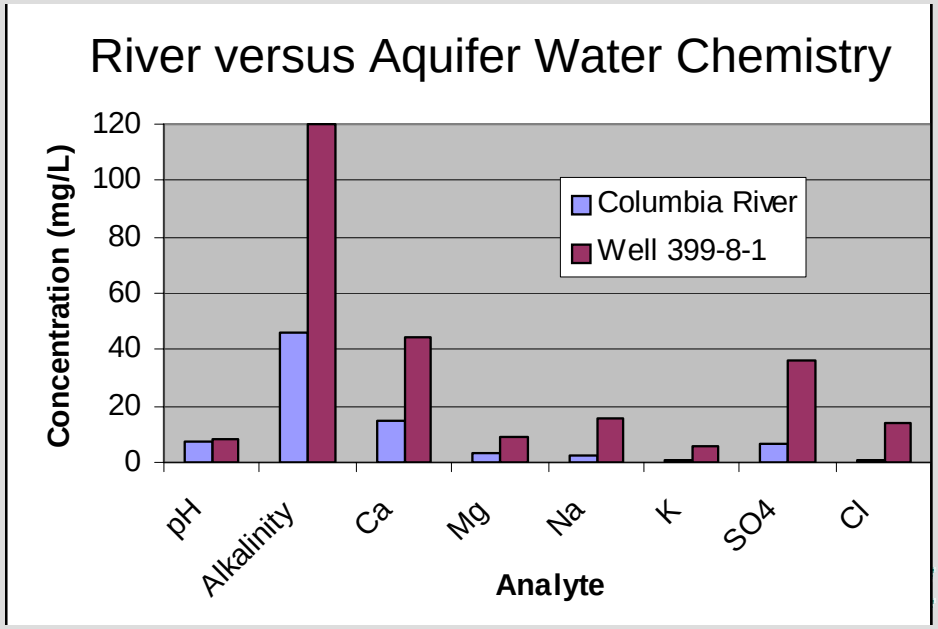
Aquifer-River Mixing Processes



- River water influx occurs during high stage
- Prolonged seasonal high stage period allows mixing in aquifer with river water
- Significant differences in solution chemistry
- 10 m/d pore velocity (Cline et al. 1985) limits residence time; potential for nonequilibrium mass transfer processes



2002 Nitrate Concentration



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Uranium Geochemistry

- Constant K_d inappropriate for temporally and spatially variable solution chemistry (e.g., uranium, pH, alkalinity)
- Nonequilibrium uranium sorption identified in column experiments with flow rates consistent with field observations
- Distinctly different sorption and desorption/dissolution rates
- Highest concentration on silt and clay size fraction

  North Process Pond capillary fringe sediments
  - 75% > 12.5 mm
  - 7% < 2.0 mm
  - 1.78% silt and clay (125 nmol/g; > 3X next highest fraction concentration)
- Key Issues:
  - leaching of uranium from cycles of saturation and drainage of contaminated vadose zone sediments
  - uranium geochemical model applicable to the full range of present and future chemical conditions in the 300 Area
  - robust relationships for sorption sites based on particle size distribution and mineralogy

300 Area Modeling Objectives

Objective

Identify the transport and fate of subsurface uranium in the 300 Area under current and likely future conditions:

- Physical and chemical processes controlling uranium mobility
  - Transport and mass transfer
    - temporally and spatially variable velocity fields
    - concentration-dependent mass transfer
  - Sorption/desorption and mineral precipitation/dissolution
    - dependent on chemical conditions (speciation, concentrations)
    - spatially variable residence times
- Processes controlling uranium fluxes at the aquifer-river interface
  - driven by hourly and seasonal variations in river stage
  - bank storage and seeps/springs
  - velocity fields in the unconfined aquifer
  - mixing zone for river and aquifer solution chemistries
- Processes controlling uranium fluxes at the aquifer – vadose zone interface
  - Percolation of recharge through uranium-contaminated vadose zone sediments
  - Saturation – desaturation of vadose zone sediments by changing water levels induced by hourly and seasonal river stage variations
    - leaching of uranium from contaminated vadose zone sediments
    - uranium uptake on clean vadose zone sediments

Requirements for Modeled Processes

- Variably saturated flow in porous media
  - 3-D vadose zone and unconfined aquifer
  - Heterogeneous subsurface materials
  - Spatially and temporally variable surface recharge
  - Dynamic river stage boundary condition
  - Temporally variable seepage face
- Multicomponent reactive transport
  - Spatially and temporally variable
    - Residence time
    - Solution chemistry
    - Solid to solution ratio
  - Rate-limited mass transfer (mobile-immobile liquid, multiregion behavior)
  - Equilibrium and Non-equilibrium Reactions
    - Aqueous complexation
    - Sorption (surface complexation, ion exchange)
    - Mineral precipitation and dissolution
  - Heterogeneous reactive surfaces

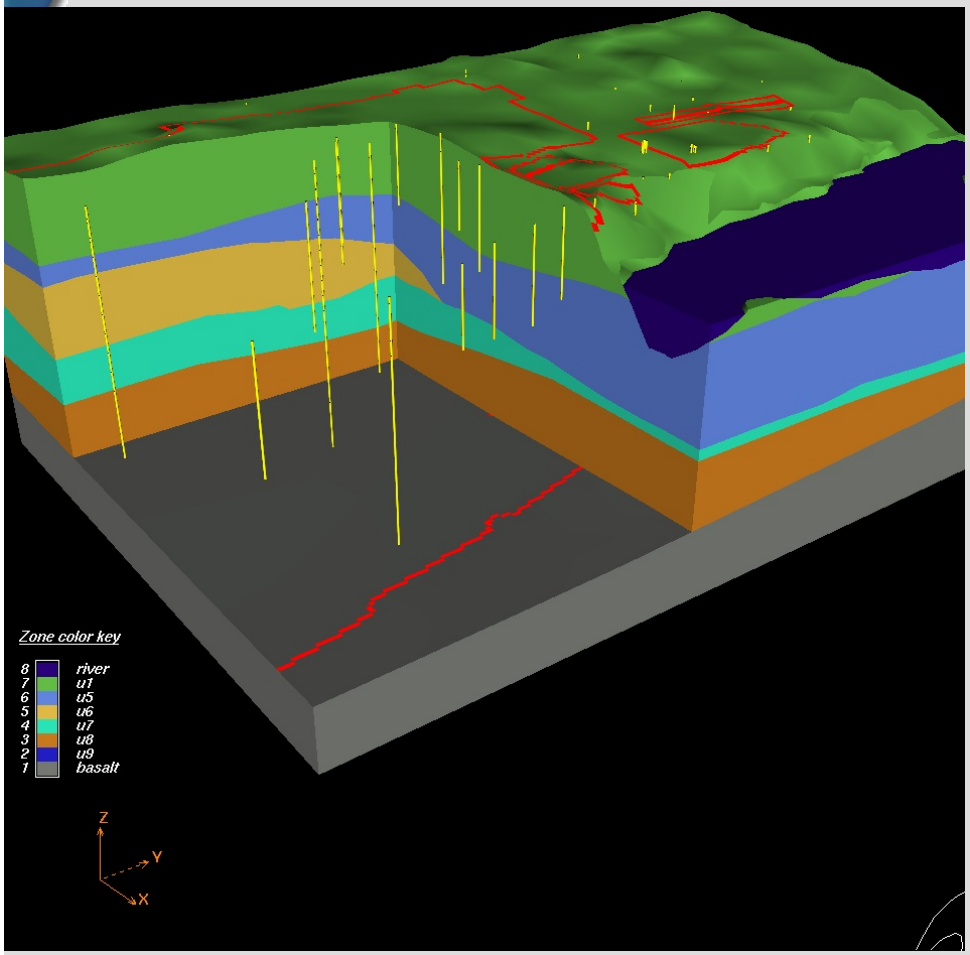
Preliminary Modeling

- 1-D Variably saturated flow and reactive transport
  - Focus: Uranium transport across Aquifer-Vadose Zone interface driven by rising and falling water table
  - Vertical columns with unsaturated uranium-contaminated sediments overlying saturated (contaminated and/or uncontaminated) sediments
  - Imbibition and drainage cycles reflecting temporal variability at principal vadose zone uranium sources (i.e., waste sites)
  - Laboratory and/or field data
- 1-D saturated flow and reactive transport
  - Focus: Uranium mobility under alternating exposure to river and aquifer solution chemistry
  - Saturated sediment exposed to alternating cycles of river water and aquifer water with uranium
  - Cycle frequency dependent on distance from river
  - Laboratory and/or field data

2-D Flow and Reactive Transport Modeling

- Vertical cross-section perpendicular to the river
- Focus: Process upscaling and coupling for uranium reactive transport driven by hourly river stage
  - Bank storage, seeps/springs, upwelling
  - Leaching of uranium from contaminated vadose zone sediments
  - Scale-up of uranium geochemistry from lab to field
- Contemporary time frame
  - No attempt to simulate historical waste loading operations
  - Availability of calibration and history matching data
- Location criteria
  - High uranium concentrations
  - Near known seeps/springs
  - Good well and data coverage
  - Extends inland beyond zone of river influence
- Existing data and assumptions for knowledge/data gaps
  - Preliminary model of uranium geochemistry
  - Large-scale hydrostratigraphic units

2-D Modeling (cont'd)



- Proposed model location
  - near Spring Location 9/11
  - near the south end of the South Process Pond
  - includes part of 307 Trenches
- Material properties
  - most recent stratigraphy
  - local variations in hydrologic properties estimated with particle size and moisture data
  - hydrologic parameter calibration using dynamic water levels and transport data
  - reactive surface area estimated from particle size and mineralogical data
- Investigate mixing driven by hourly river stage fluctuations
  - assess need for mixing model to be used with longer (e.g., monthly) time steps
- Investigate dynamics of riverbank storage, spring formation, and fluxes across aquifer – river interface
- Investigate release of uranium from contaminated vadose zone sediments
  - applicability of laboratory derived parameters
  - calibration of field scale parameters

3-D Flow and Reactive Transport

- Build on new knowledge
  - Characterization and Monitoring Programs
  - Geochemical studies
  - Updated process models and parameterizations from 1-D, and 2-D investigations
  - NRC 3-D model development: grids, material property distributions, initial and boundary conditions
- Third dimension will expand 2-D domain north and south
  - 300 Area uranium plume
  - Major uranium source zones (process trenches)
  - Known uranium seeps and springs
- Focus
  - Vadose zone uranium inventory
  - 300 Area uranium flux across aquifer-river interface driven by hourly flow
  - Long-term vadose zone uranium sources driven by rising and falling water table
  - Plume migration and attenuation
  - History-matching to a field campaign

Knowledge Gaps and Data Needs

- Uranium transport behavior (laboratory and field)
  - Mobility of vadose zone uranium under cycles of saturation and desaturation
  - Mobility of uranium in the aquifer under cycles of exposure to distinctly different river and aquifer water chemistries
  - Mobility of uranium in different aquifer materials
- Uranium inventory in the vadose zone
  - Mass and spatial extent beneath waste disposal sites
  - Vertical distribution with depth especially near capillary fringe
  - Chemical forms
    - Speciation
    - Partitioning between liquid and solid phases
    - Association/Correlation with particle size and specific mineral surfaces
- Uranium plume(s) in the aquifer:
  - Concentration distribution and spatial extent
  - Vertical distribution

Knowledge Gaps and Data Needs (cont'd)

- Physical process parameters
  - Large-scale
    - spatial distribution of material properties consistent with water table and transport response to river stage fluctuation
      - Flow: hydraulic conductivity, porosity, pressure-saturation-relative permeability functions
      - Transport: dispersivity
    - surface recharge
      - Spatially and temporally variable
      - Current and future land use
  - Small-scale
    - Preferential pathways for uranium transport in unconfined aquifer
    - Discharge of uranium into the river system including likely areas of upwelling into the riverbed
- History matching data for model confirmation
  - Short periods of high frequency data collection (hourly) for near-river and in-river wells
  - Daily to seasonal data collection frequency for selected set of wells
  - Multilevel sampling in aquifer and wetting interval of vadose zone