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# Field Test of Vapor-Phase Ammonia Injection for Vadose Zone Remediation of Uranium

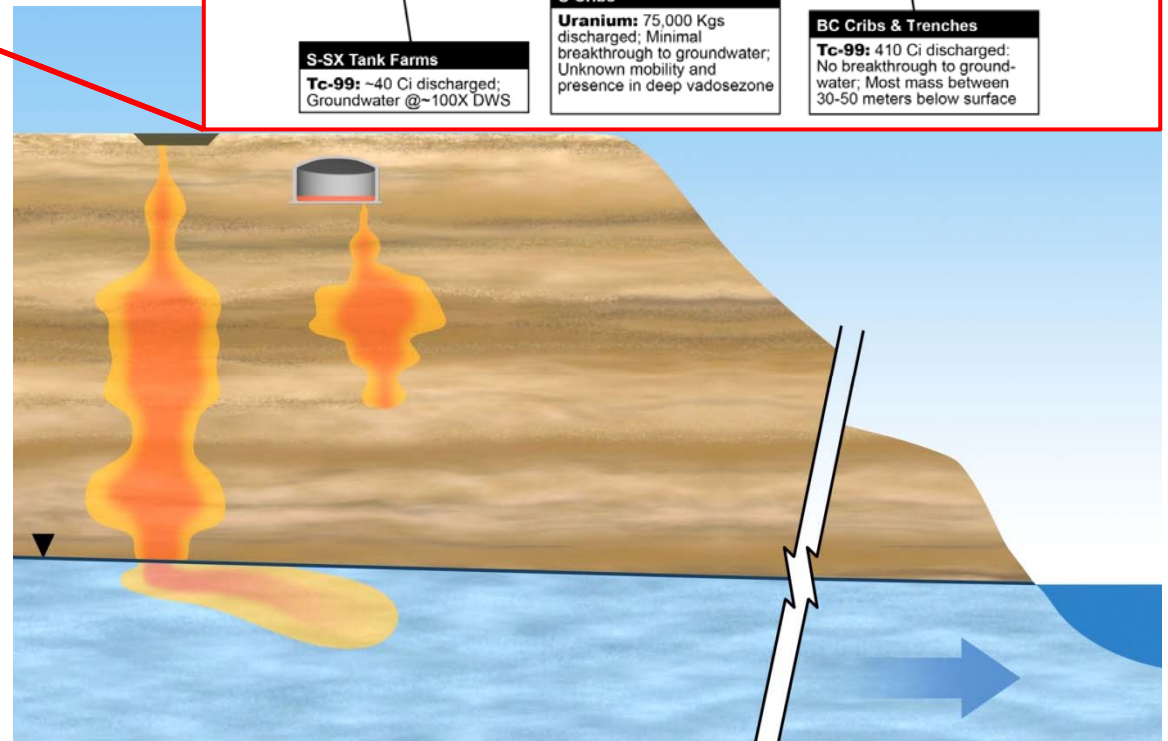
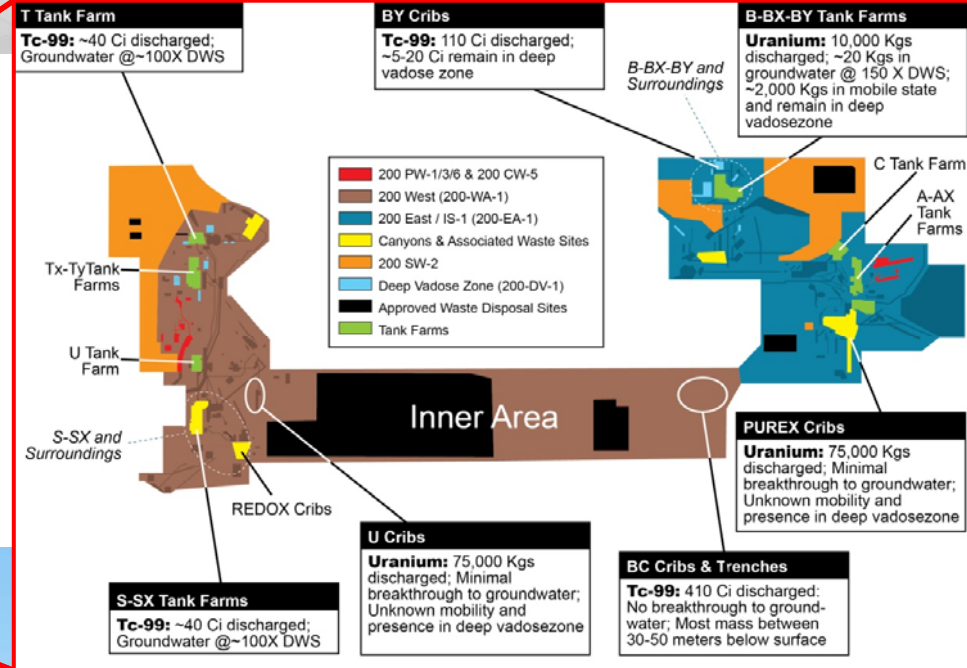
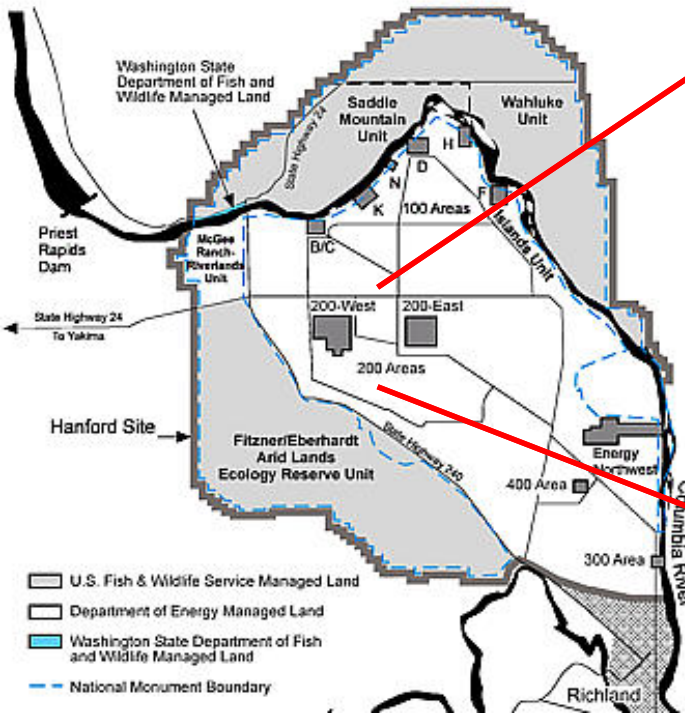
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- ▶ Large inventory of contaminants in the vadose zone at Hanford: concern as a potential source of future groundwater contamination
- ▶ Efforts underway to develop in situ vadose zone remediation for radionuclide contaminants
- ▶ Uranium is of concern because of its large inventory and mobility

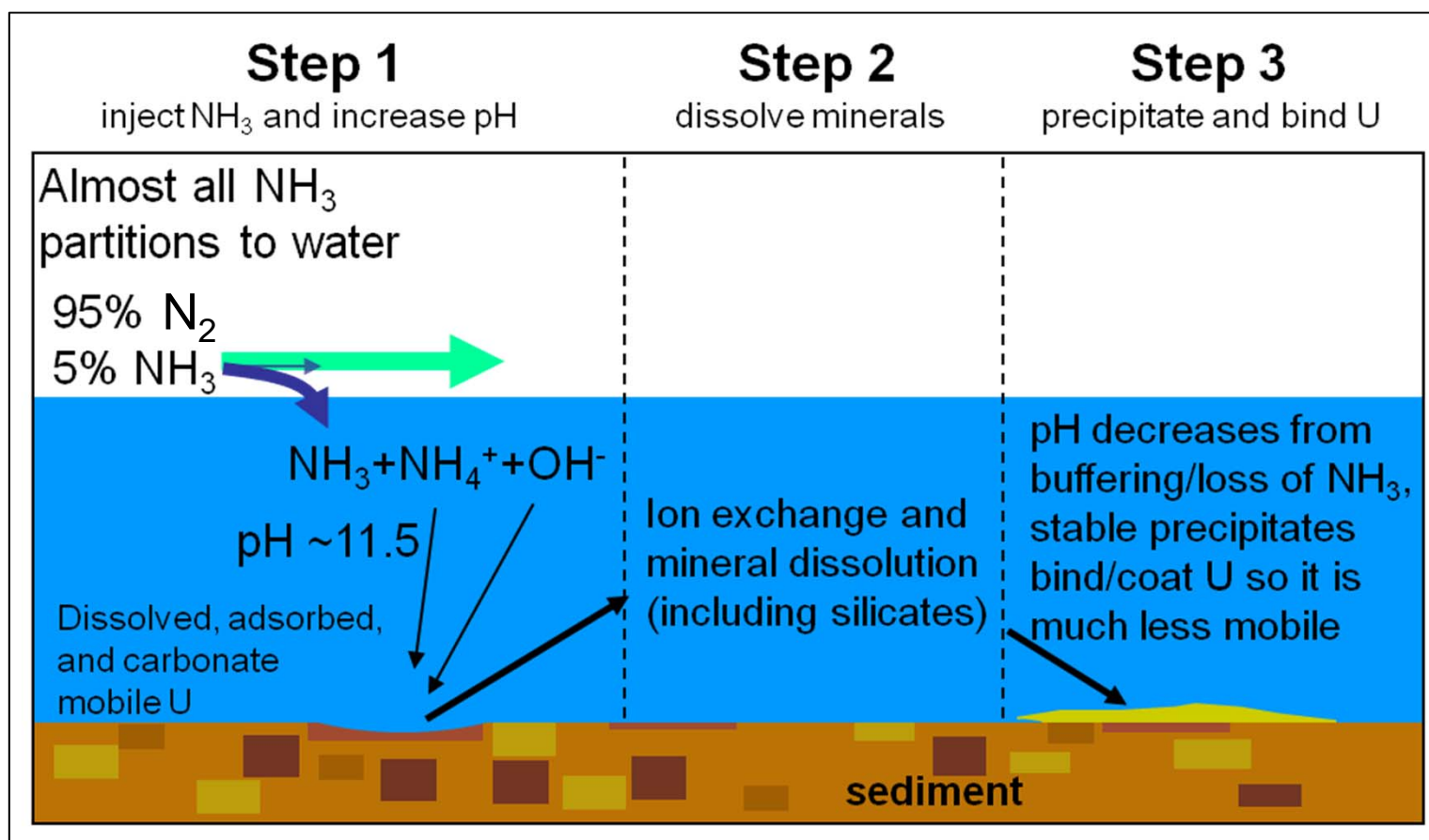
# Introduction



- ▶ Investigating geochemical manipulation to change the subsurface conditions in a way that slows downward migration of the contaminants
  - Focus of efforts is use of amendments delivered in the gas-phase
  
- ▶ Development from concept to field application
  - Concept: Introduction of caustic waste fluids to the subsurface dissolves part of the sediment. Subsequent precipitation can bind or coat contaminants and render them less mobile.

# Ammonia Treatment

- ▶ Ammonia can be delivered in the gas phase and creates caustic conditions in the pore water.

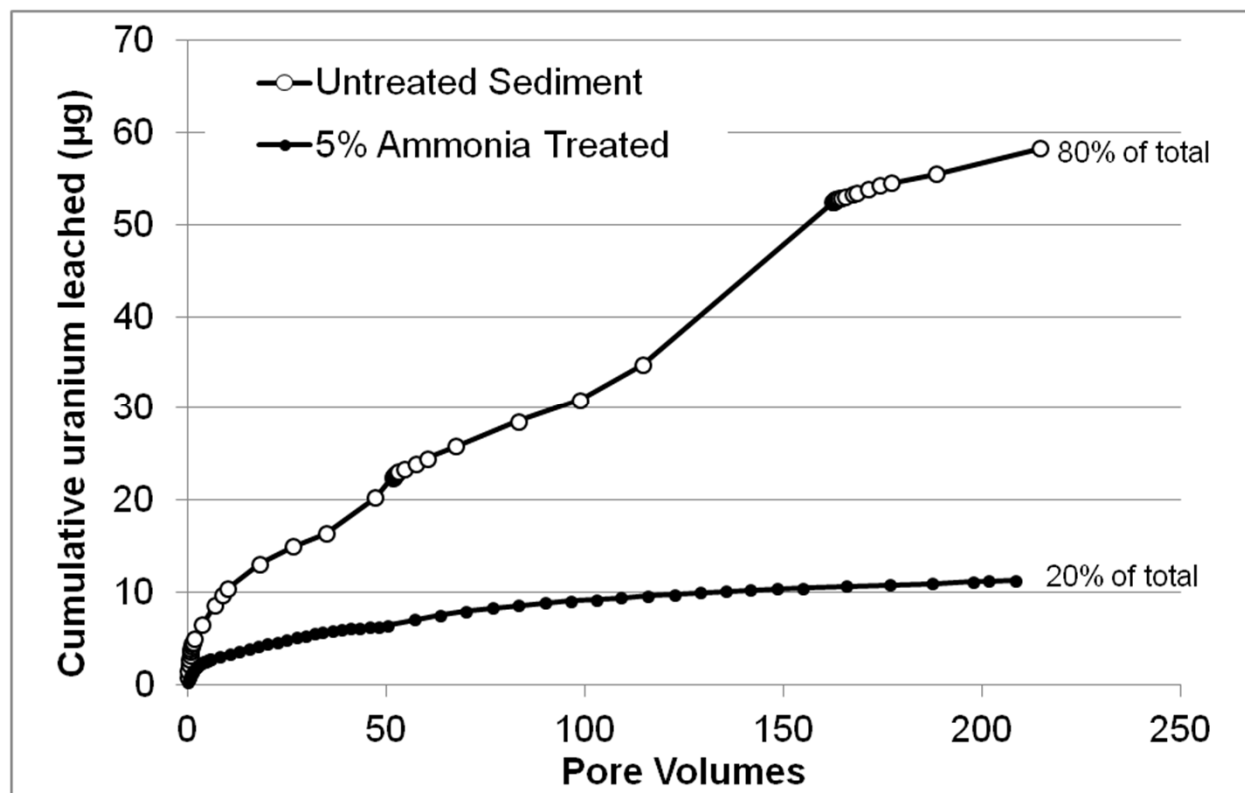
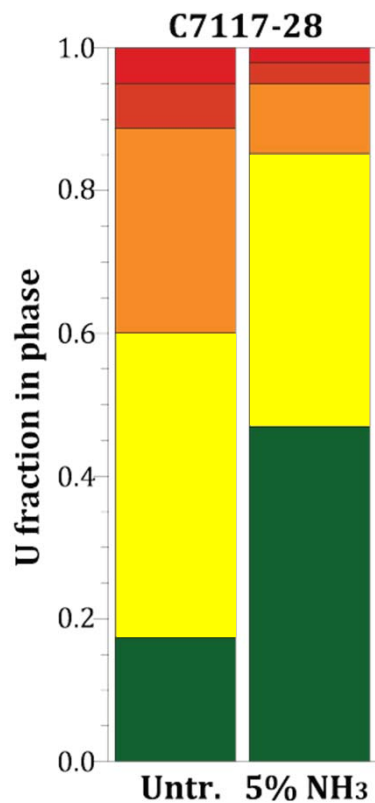


# Evaluating Treatment Effectiveness

## ▶ Sequential extraction method

- Groundwater (mobile in pore water)
- Ion exchange (mobile, sorbed)
- pH 5 acetate (moderately mobile, carbonate rind)
- pH 2.3 acetic acid for 1 week (slow release, carbonate)
- 8M Nitric acid at 95C (functionally immobile, total)

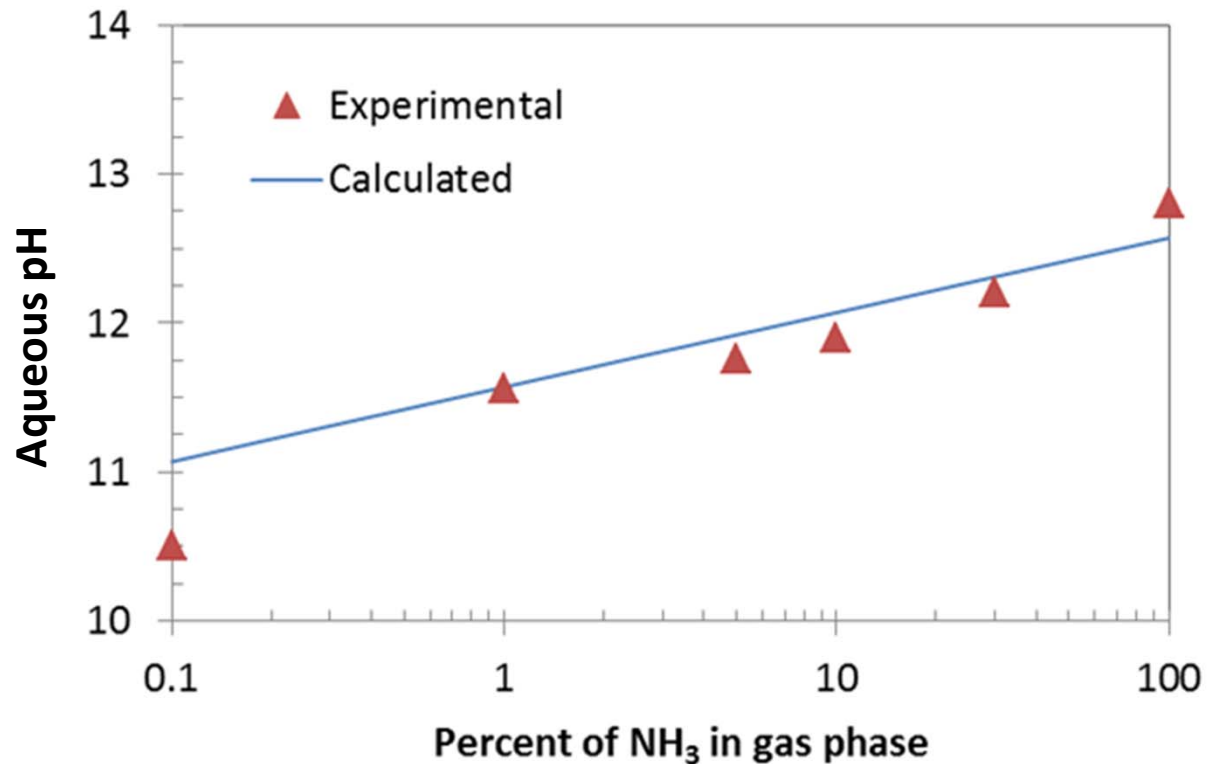
# Sequential Extraction and Leaching Results





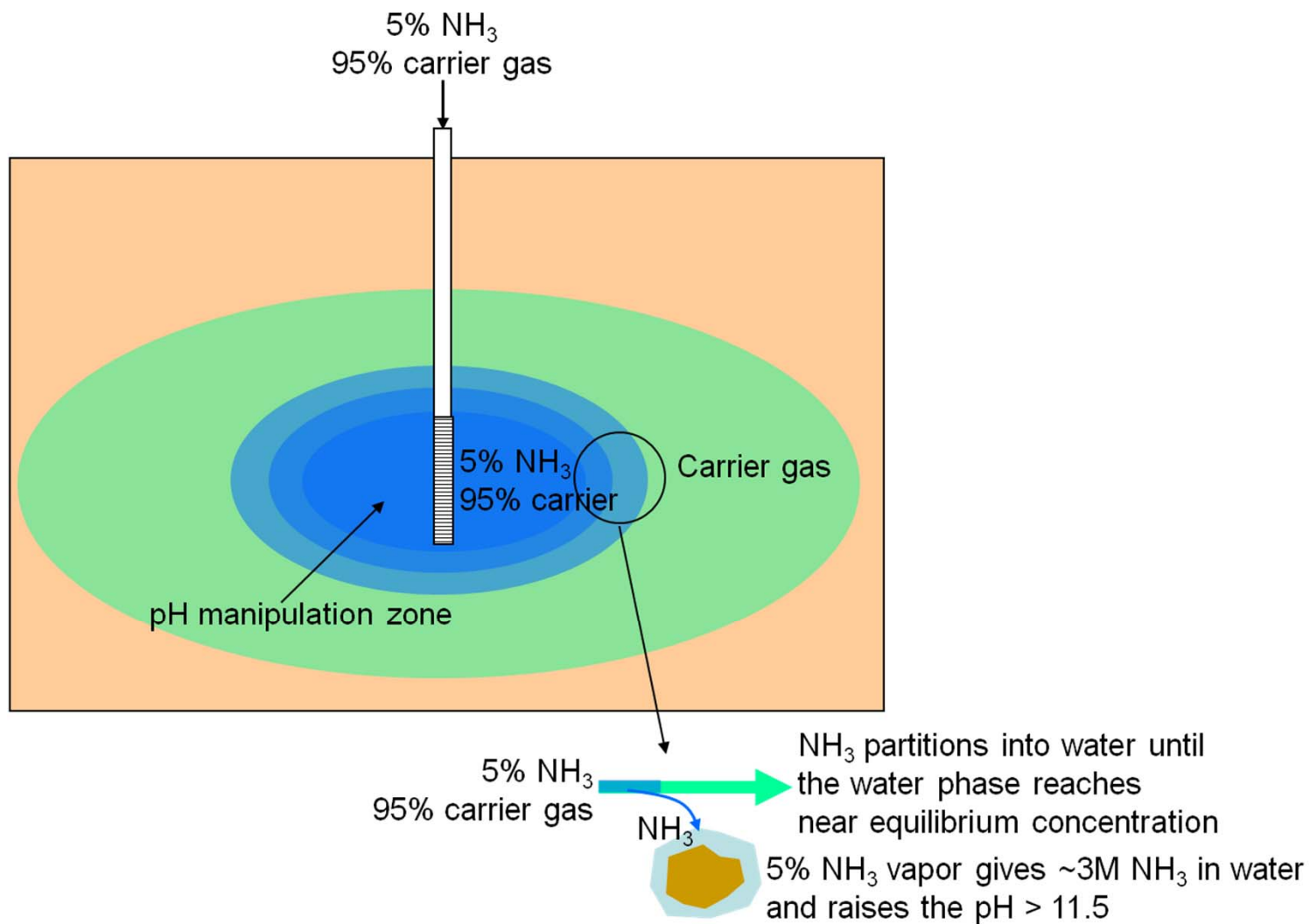
# Ammonia Partitioning into Water

- ▶ Partitioning is reasonably well predicted by Henry's Law such that field design calculations can be developed



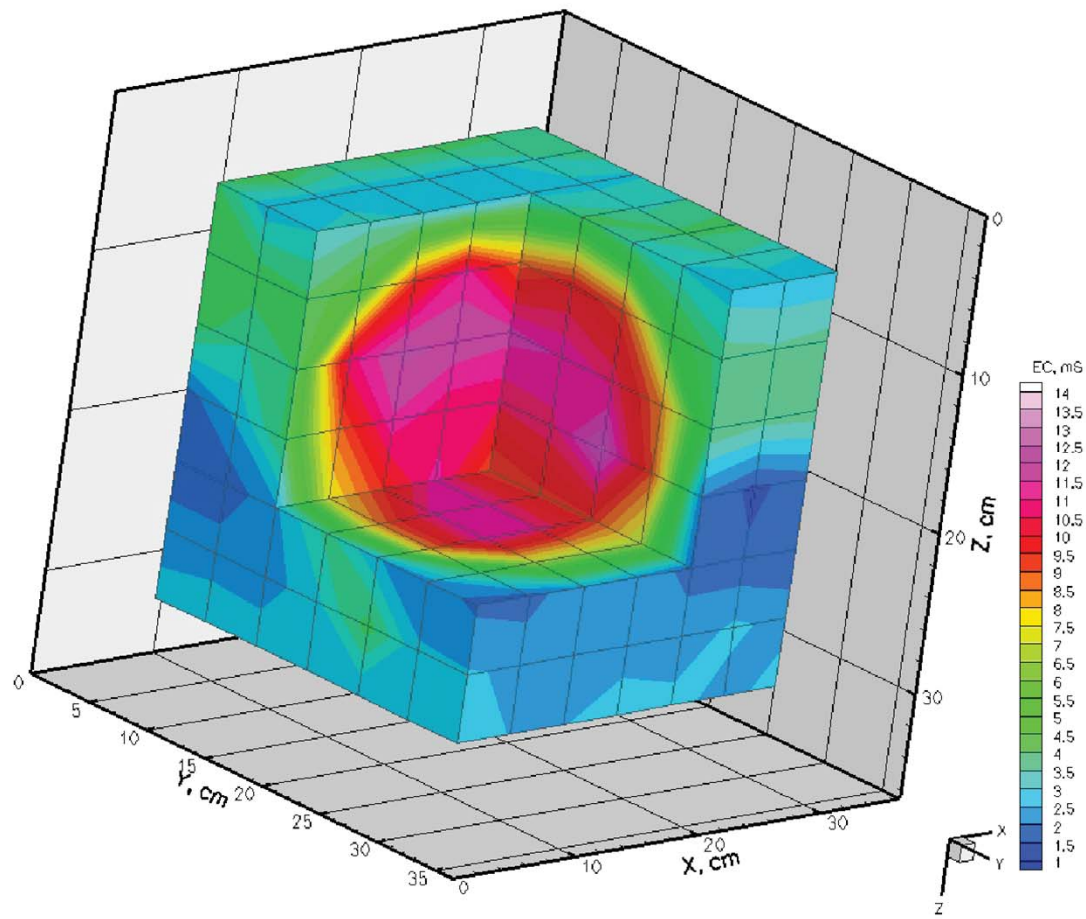


# Field Design



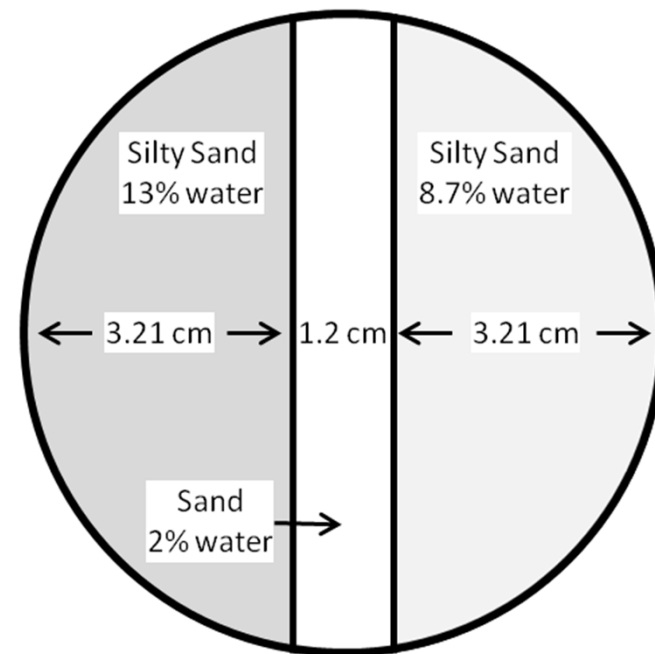
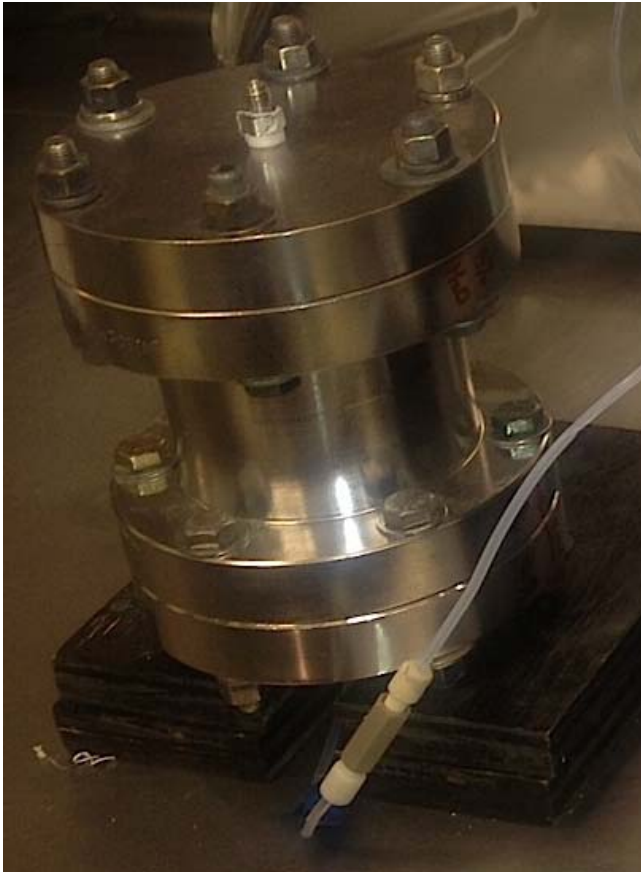
# Ammonia Delivery

- ▶ Ammonia injected into a cube of sediment



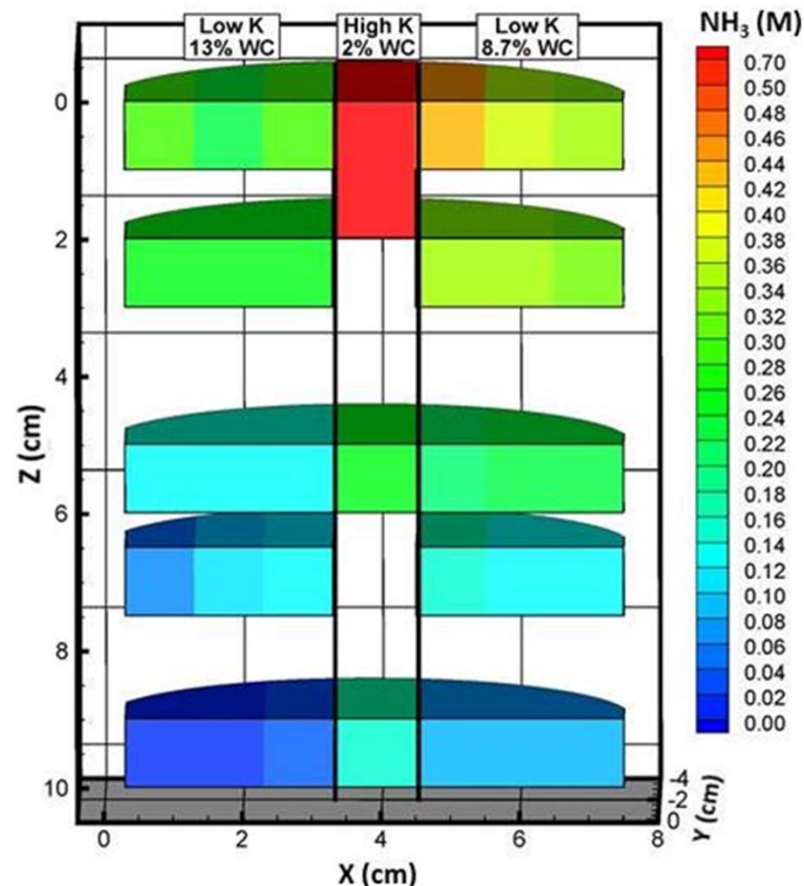
# Treatment of Low Permeability Zones

- ▶ Plan view of packing for a large soil column test (10 cm length)



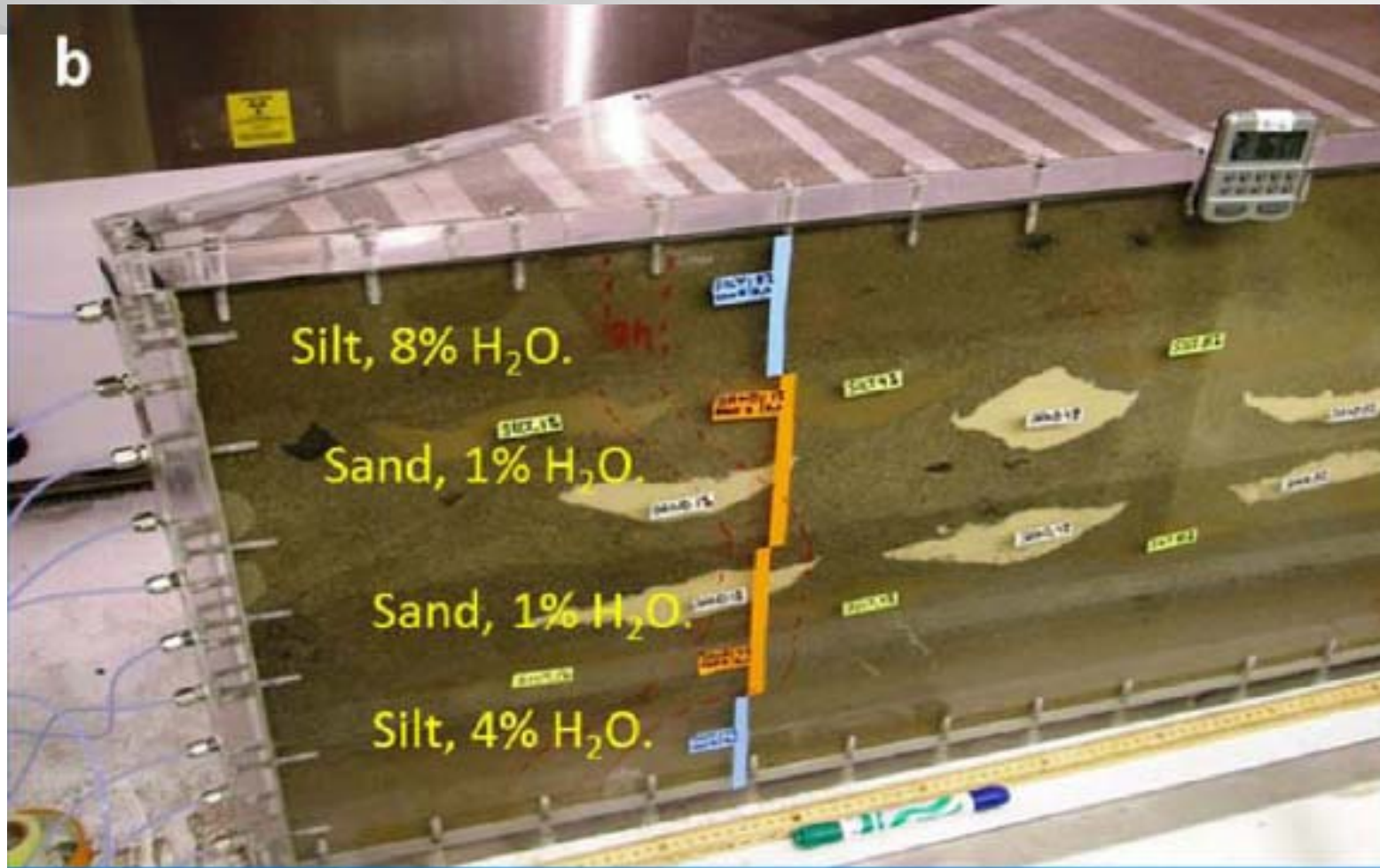
# Vadose Zone Considerations

- ▶ Diffusion of a 0.1 M ammonia pore-water concentration front ( $\text{pH} > 11$ ) from a 5% ammonia gas boundary
  - 5 cm/week, and 8.7% moisture silty sand
  - 3.4 cm/week for 13% moisture silty sand





# Laboratory Injection



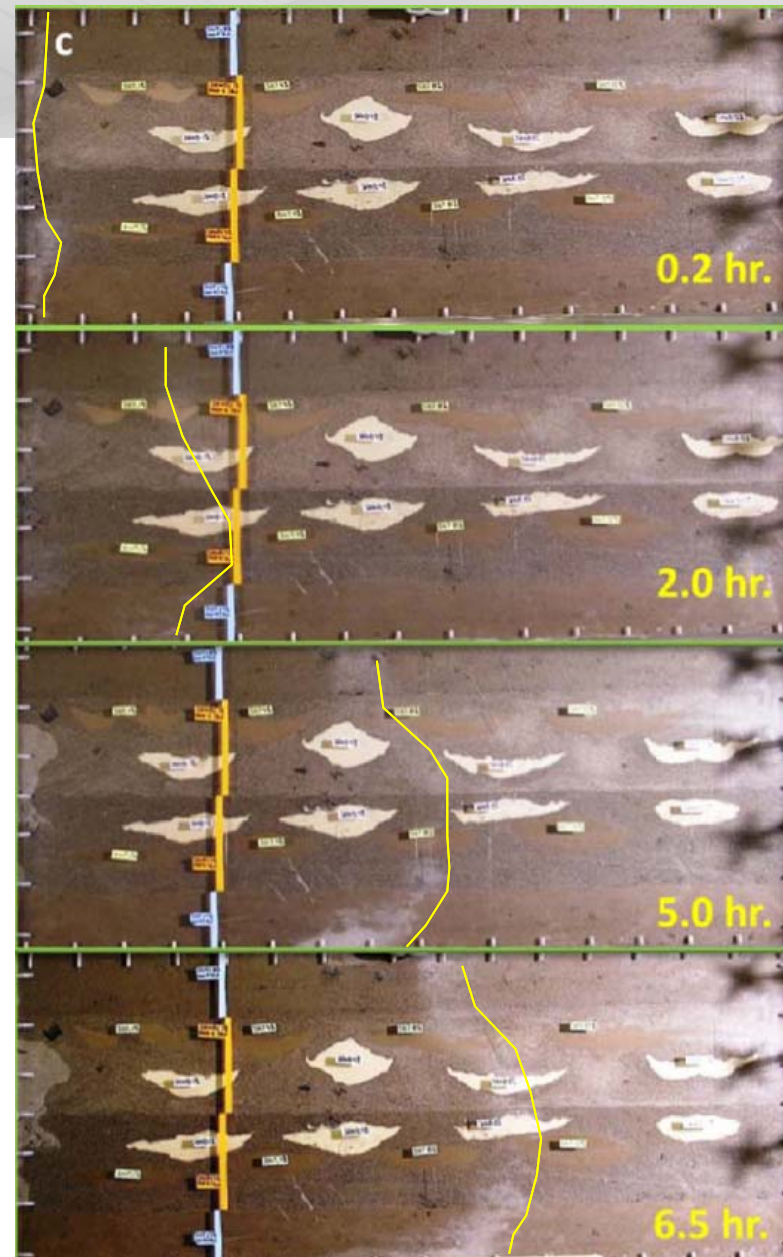
# Laboratory Injection



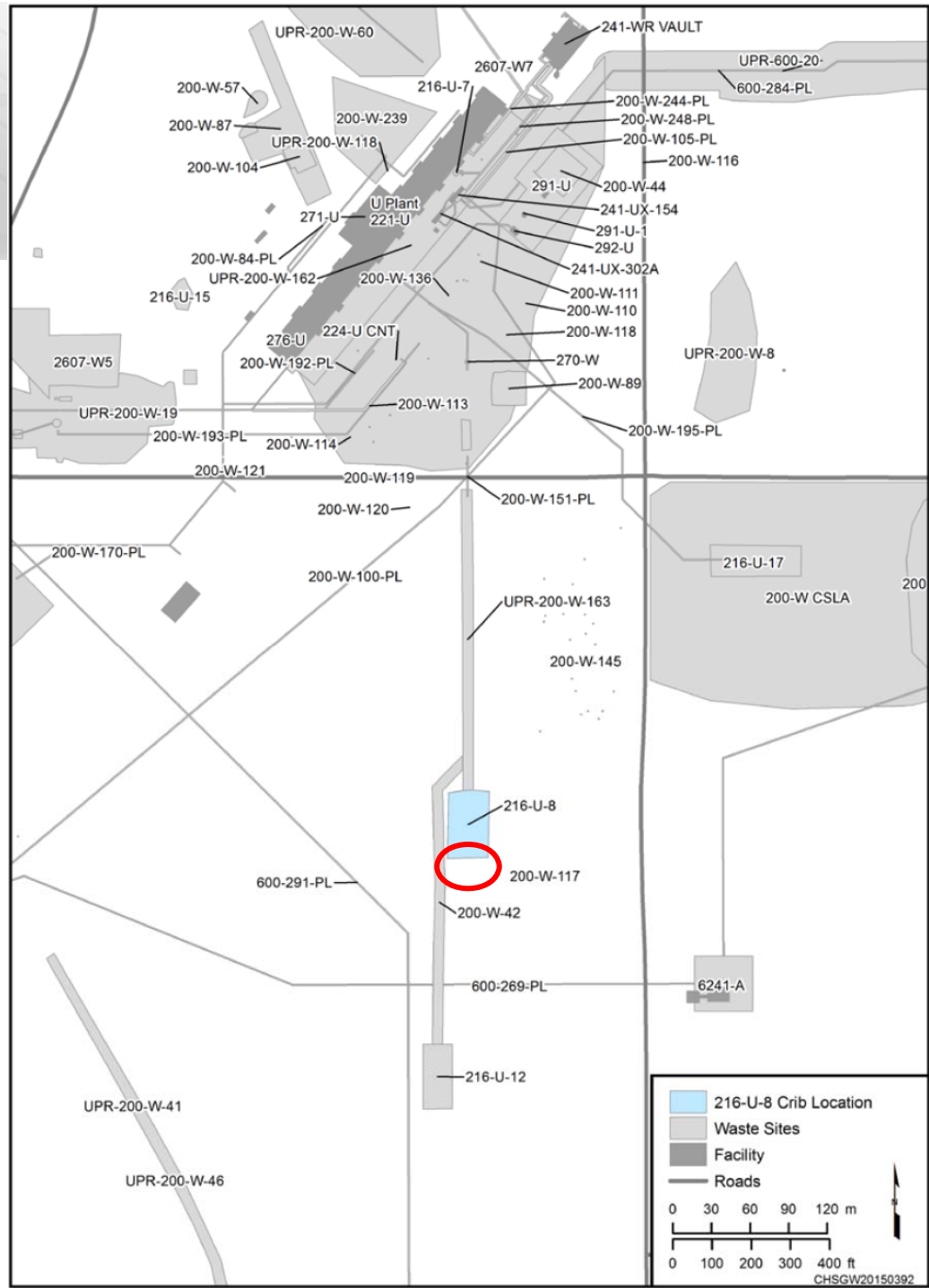
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- ▶ Injection across permeability contrasts showed relatively even movement of the injection front.
- ▶ Post injection analysis showed ammonia distribution into fine sand and silt lenses

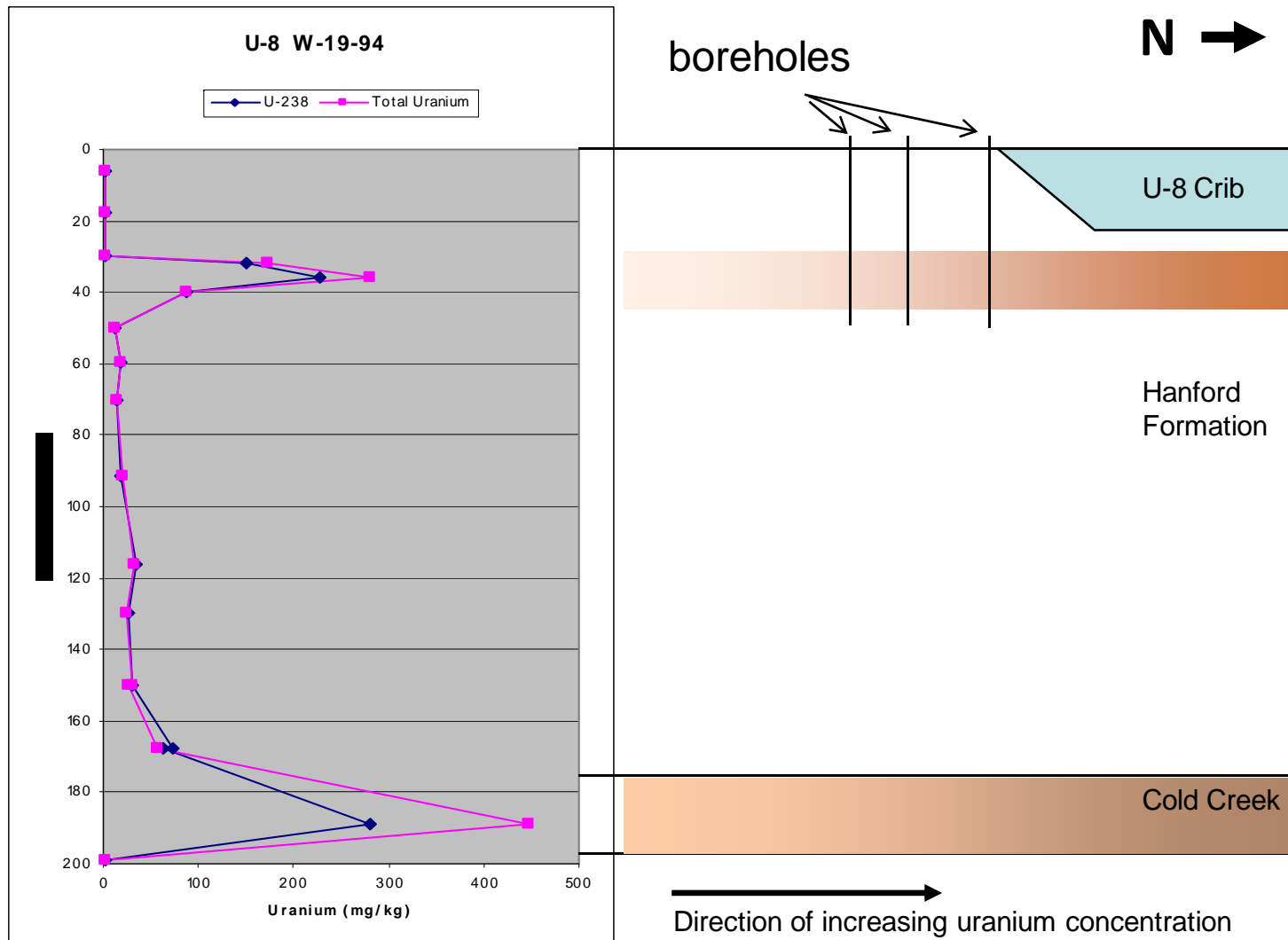


# Test Location: 216-U-8 Crib





# Uranium Distribution and Target Test Zone at the 216-U-8 Site



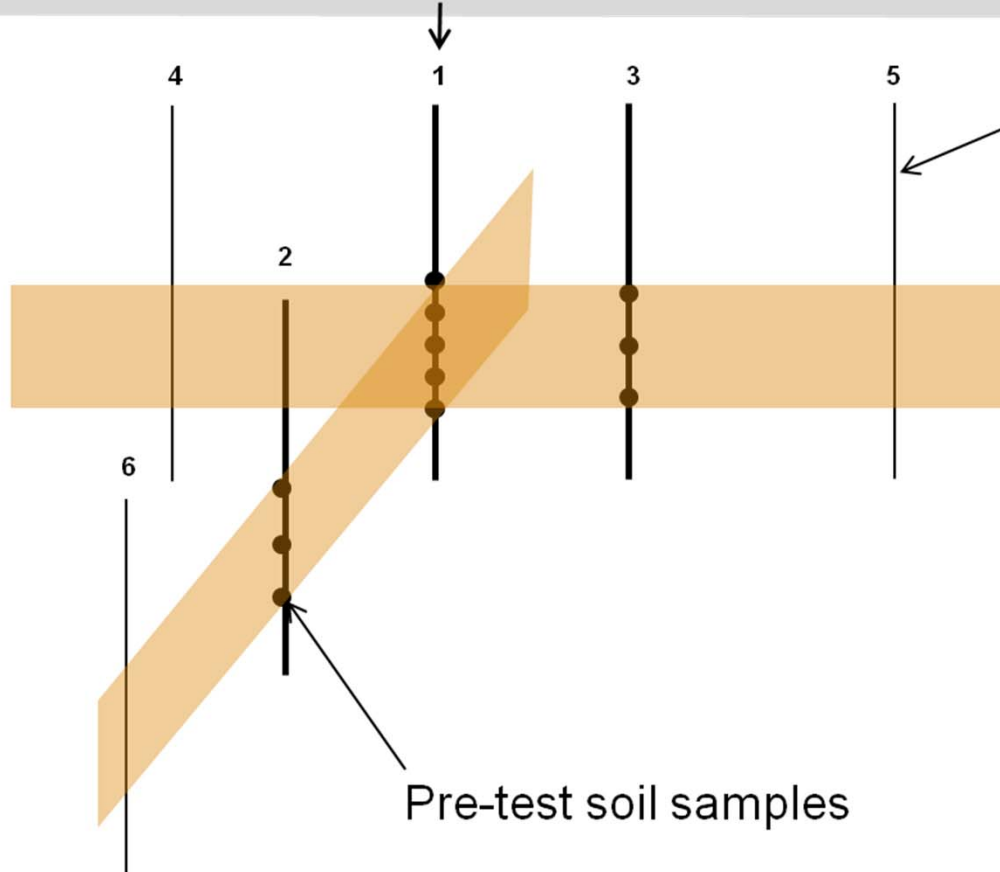
# Site Well/Borehole Layout



U-8 Crib

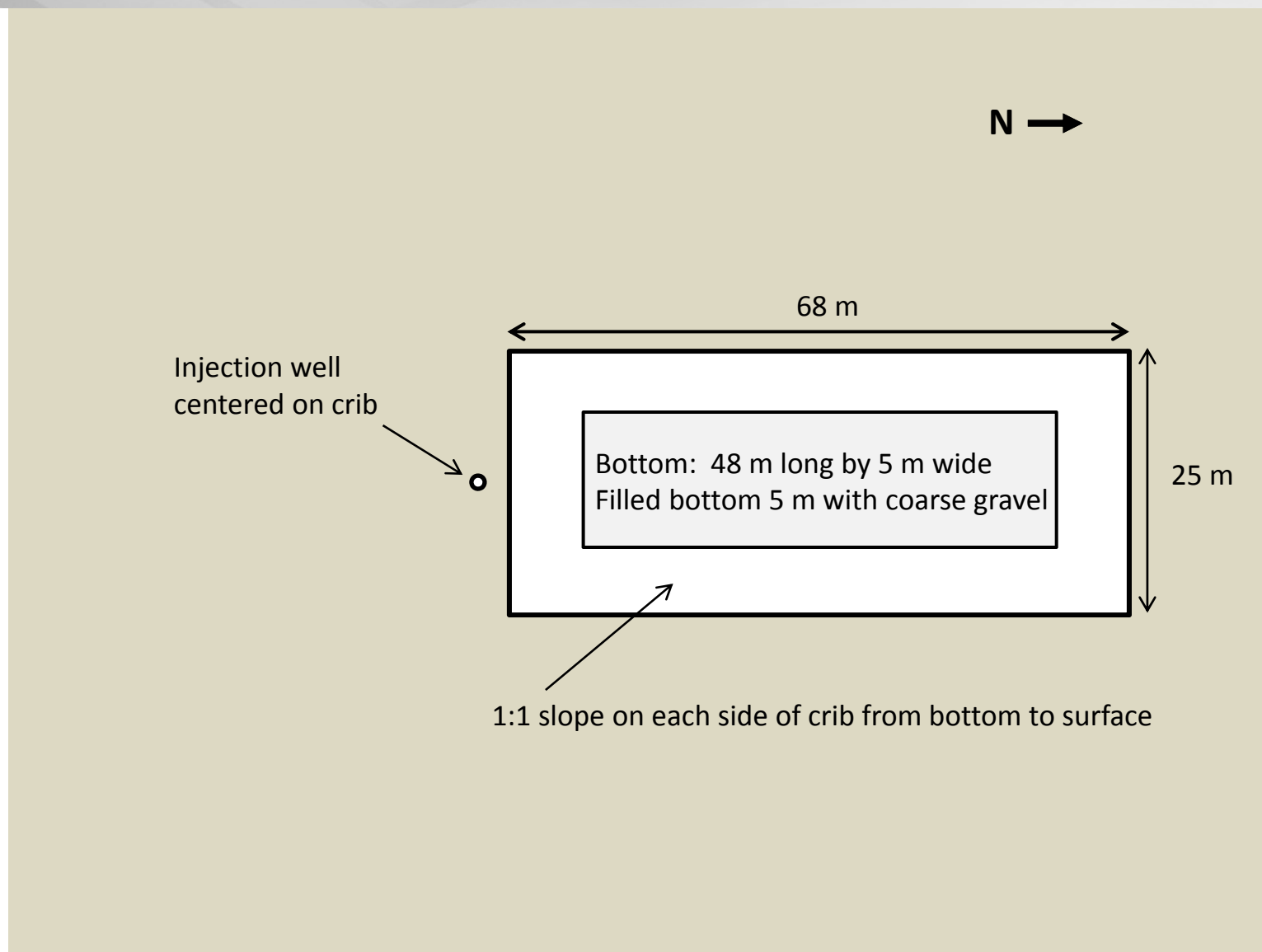
Injection Well

Instrumented boreholes

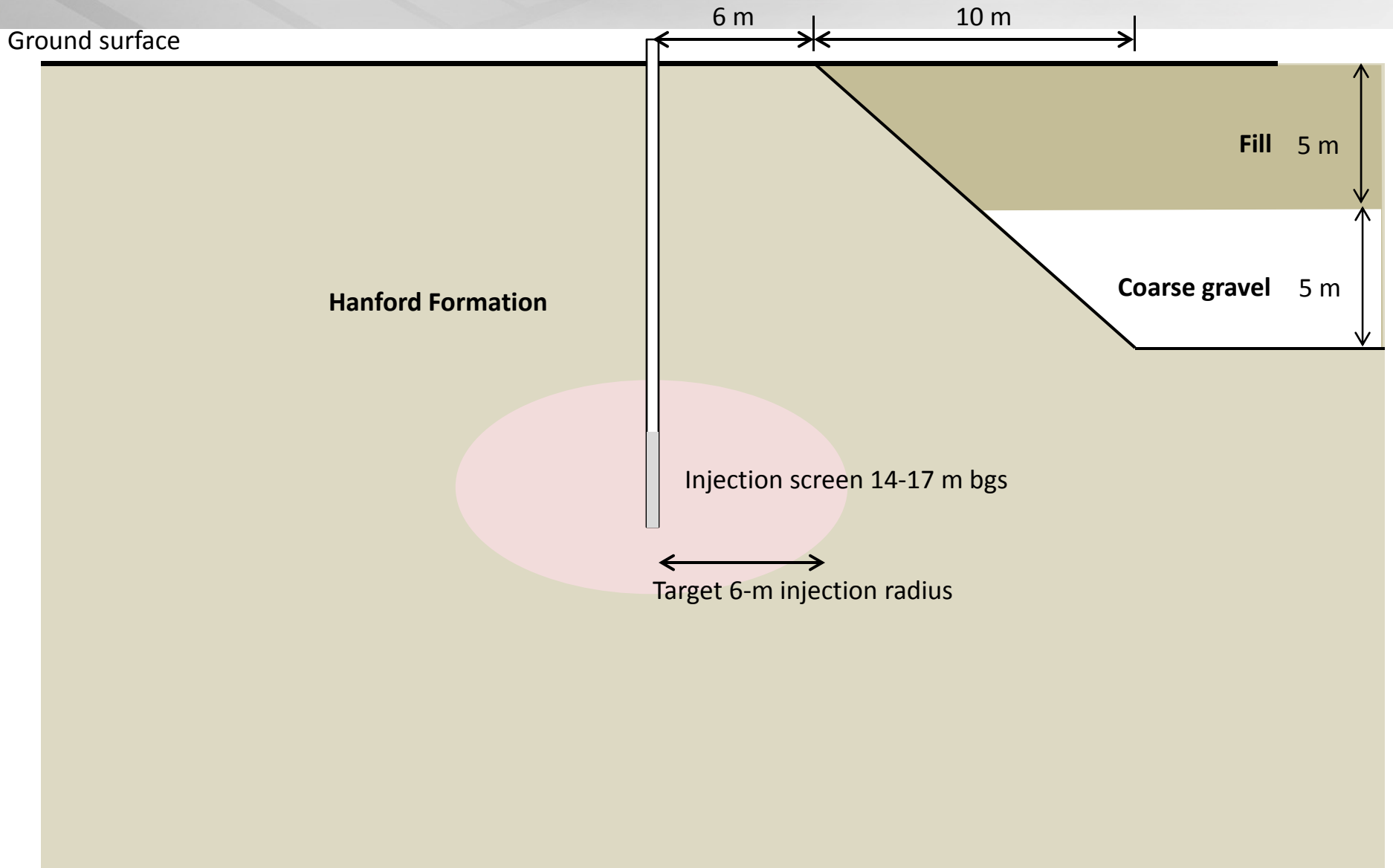


Pre-test soil samples

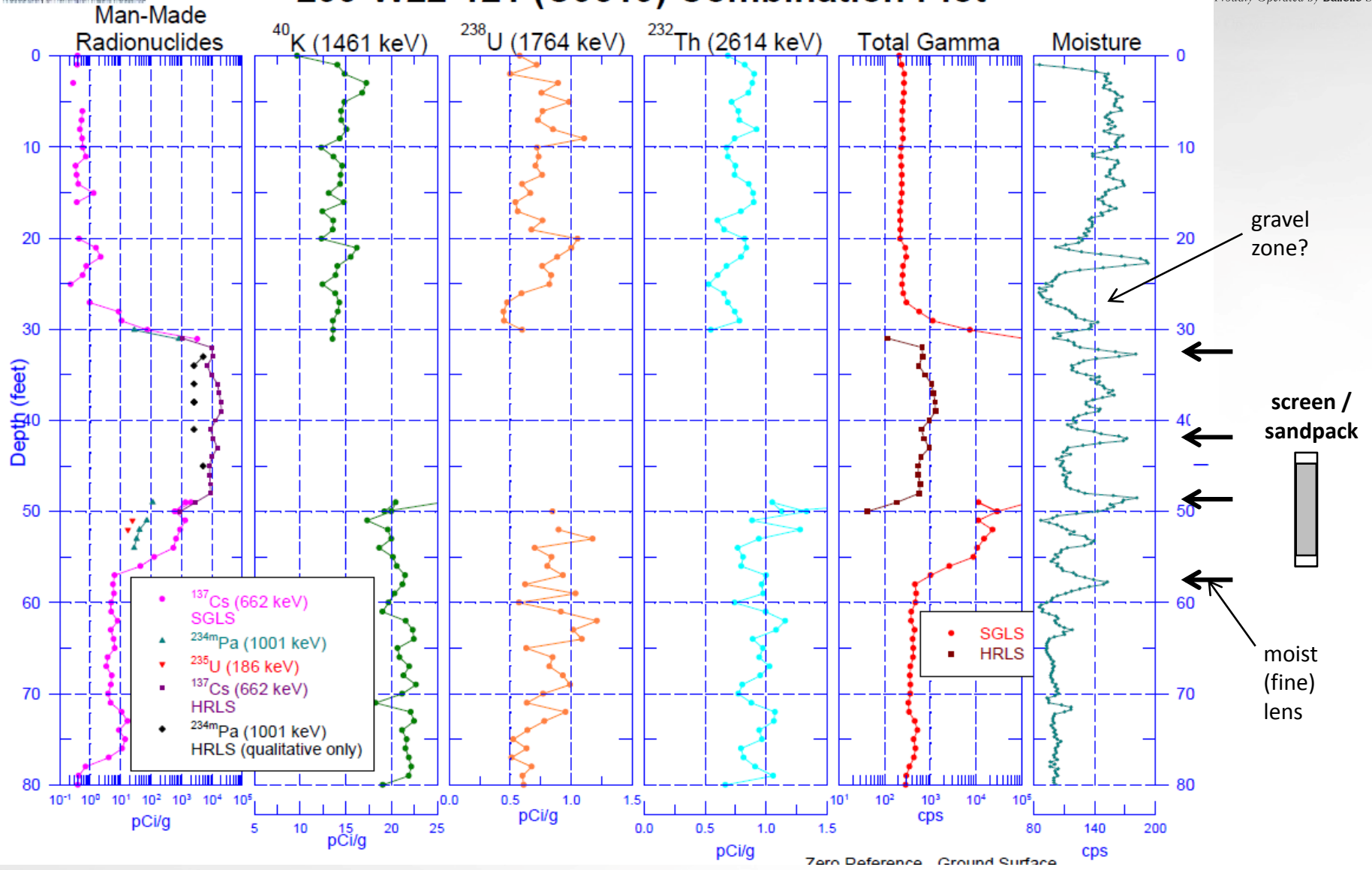
# Plan View



# Cross Section



# 299-W22-121 (C9519) Combination Plot



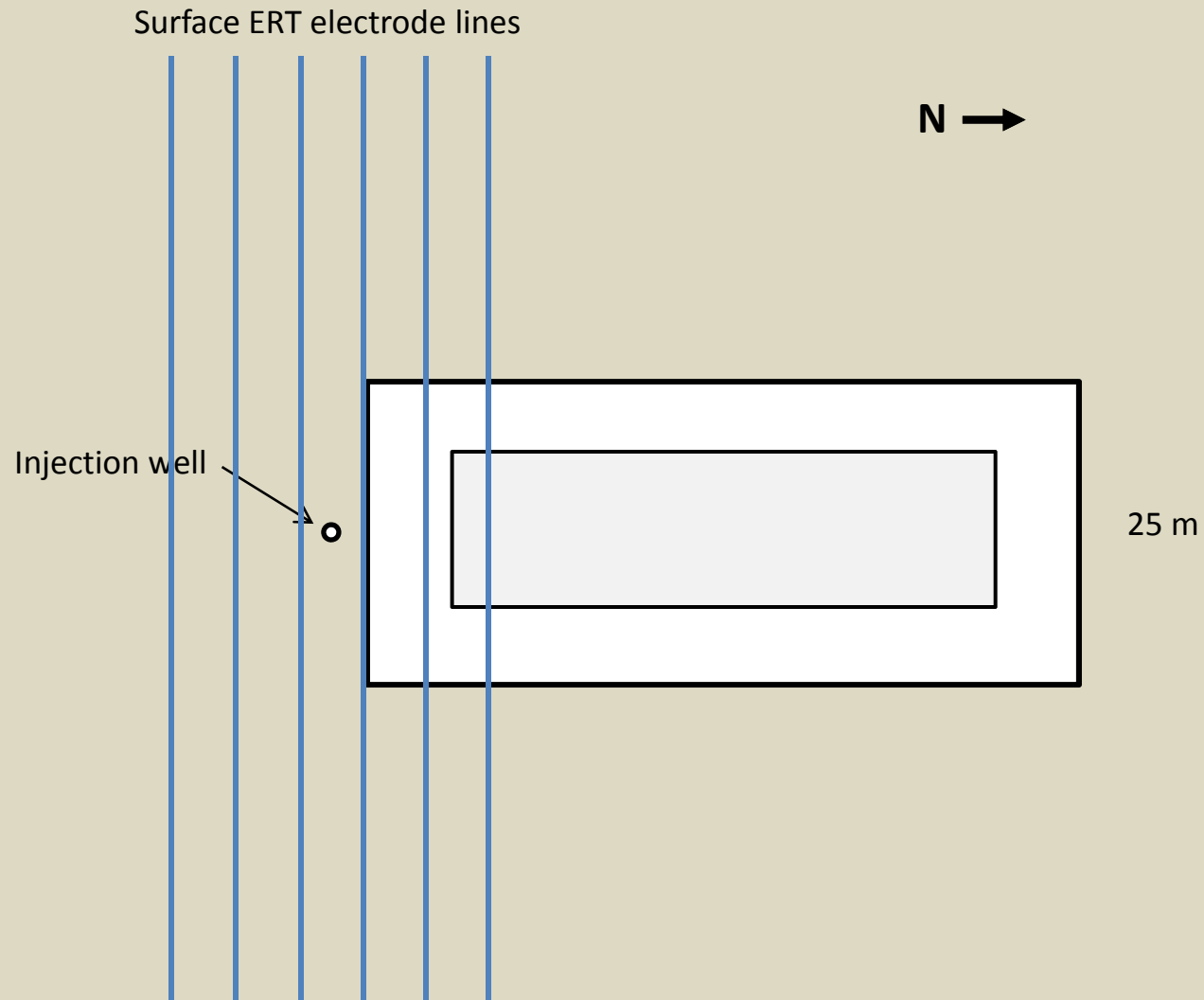
- ▶ Ground Surface Ammonia Monitoring
  - Ammonia trailer
  - All piping joints
  - Perimeter/area monitors at ground surface
  - Subsurface gas sampling ports
  
- ▶ Injection Monitoring
  - Electrical Resistivity
  - Temperature
  - Subsurface gas samples

# Surface Electrical Resistivity Tomography



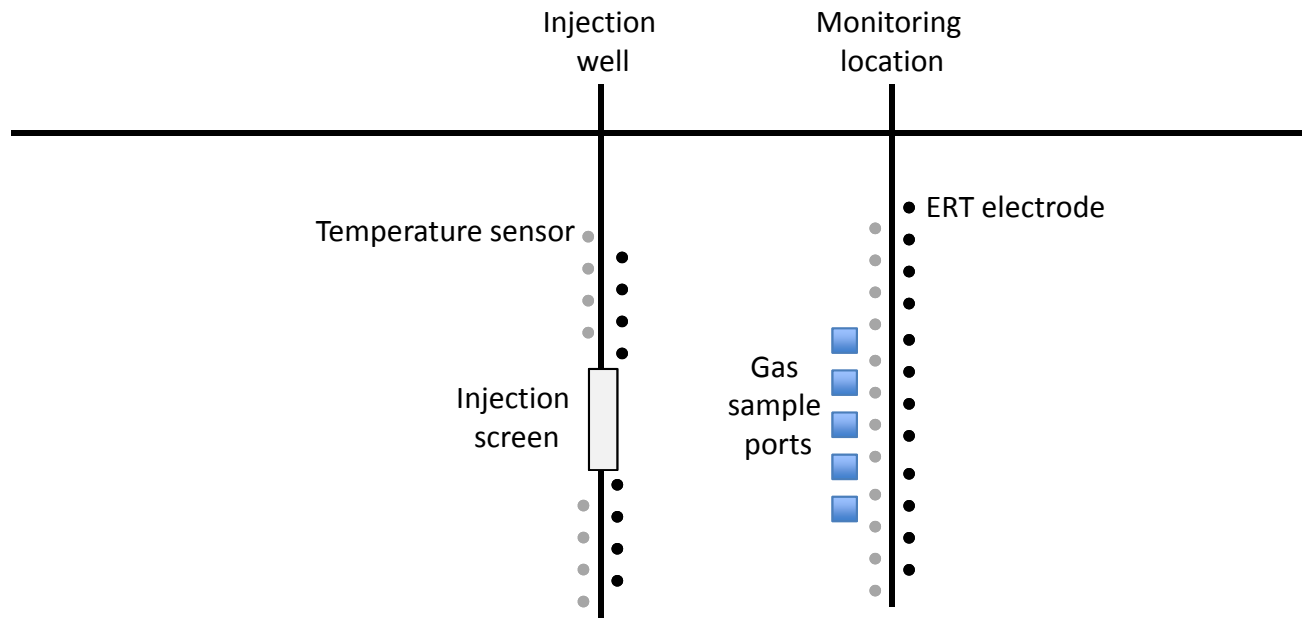
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- ▶ Cross-hole ERT
- ▶ Monitoring for lateral and vertical movement



# Test Objectives

- ▶ Determine design and operational parameters
- ▶ Demonstrate field-scale treatment
- ▶ Demonstrate field-scale equipment
- ▶ Collect sufficient information to support consideration of ammonia treatment for a feasibility study

# Test Design Issues

- ▶ Ammonia stock is a liquefied gas
  - Pressure depends on temperature (controlled)
  - Cooling with conversion to gas
  - Mass-flow controller for gas-phase mixing with nitrogen gas
- ▶ Need anhydrous conditions
  - Ammonia strongly partitions into water
  - Desiccation will occur near injection well
- ▶ Ammonia smell recognized well below hazard level
  - Good warning
  - Personnel concerns

- ▶ Equipment compatibility with ammonia
- ▶ Ammonia “reaction” time
  - Pore water concentrations drop over first week or so after injection ceases
  - Temporary interruptions of injection
    - Hours to a few days – no impact
    - Week – may “re-treat” areas already treated
  - Full reaction time for precipitation is months to a year with longer as better
    - In vadose zone “reaction time” is not critical because transport rate is slow
- ▶ Ammonia will follow carrier gas flow pattern but be slowed and diffuse more due to interaction with water
  - Still need to consider short-circuit flow paths

- ▶ Field equipment installed and ready for injection
- ▶ Administrative hold to address concerns for use of ammonia
  - Hazards review board
  - Concern for surrounding activities
    - Timing of activity

# Conclusions

- ▶ Vadose zone remediation is aimed at decreasing the contaminant flux from the vadose zone to the groundwater
- ▶ Geochemical manipulation with ammonia creates low-solubility precipitates that are effective in reducing uranium mobility
  - Not sensitive to re-oxidation
  - Favorable delivery properties for the vadose zone
- ▶ Use of ammonia must consider hazards and appropriate controls

# References

- ▶ Department of Energy. 2008. “Deep Vadose Zone Treatability Test Plan for the Hanford Central Plateau.” DOE/RL-2007-56, Revision 0, U.S. DOE Richland Office, Richland, WA.
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- ▶ Szecsody, J.E., M.J. Truex, L. Zhong, T.C. Johnson, N.P. Qafoku, M.D. Williams, J.W. Greenwood, E.L. Wallin, J.D. Bargar, and D.K. Faurie. 2012. Geochemical and Geophysical Changes During NH<sub>3</sub> Gas Treatment of Vadose Zone Sediments for Uranium Remediation. *Vadose Zone J.* 11(4) doi: 10.2136/vzj2011.0158.
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