Integrated Evaluation of Leaching Processes for Environmental Assessment

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NIEHS Workshop on Arsenic Leaching From Drinking Water Treatment Residues
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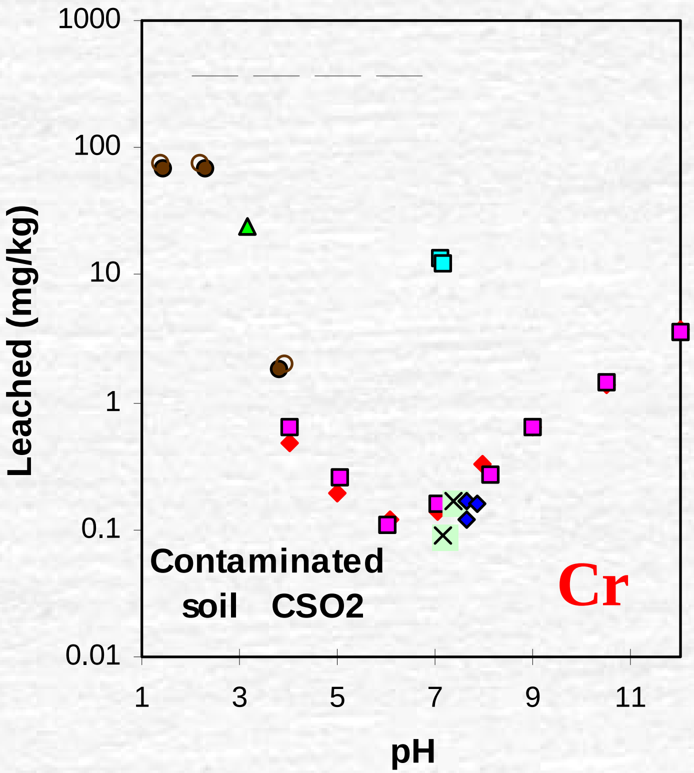
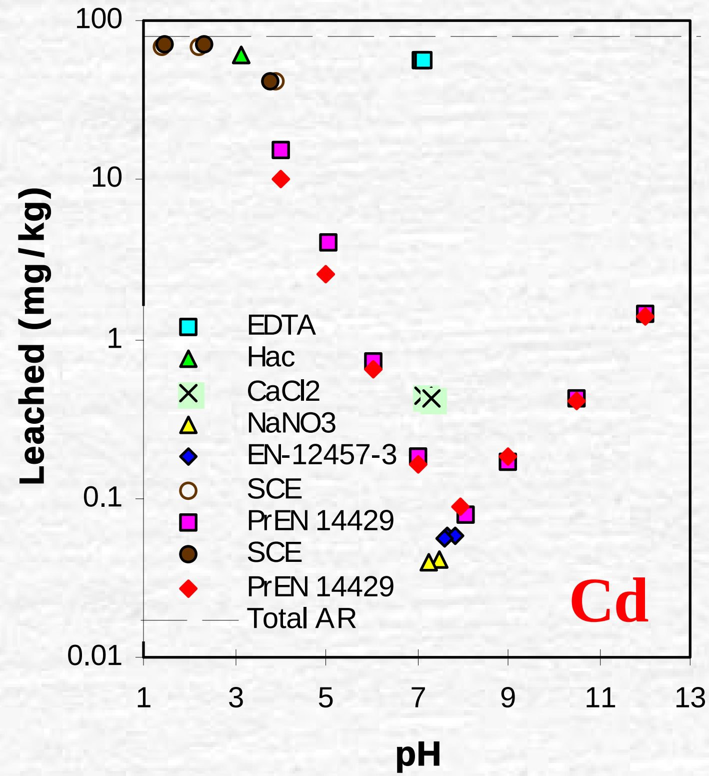
Comparison of Different Leaching Tests

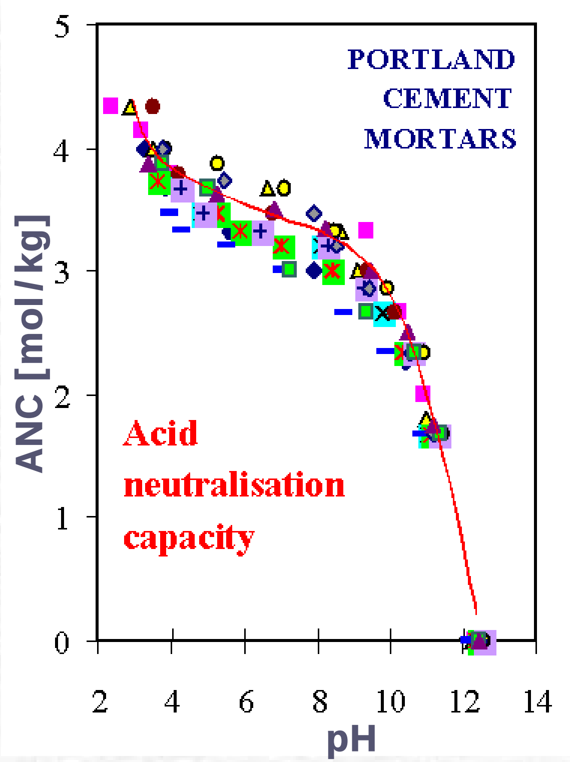
Contaminated Soil

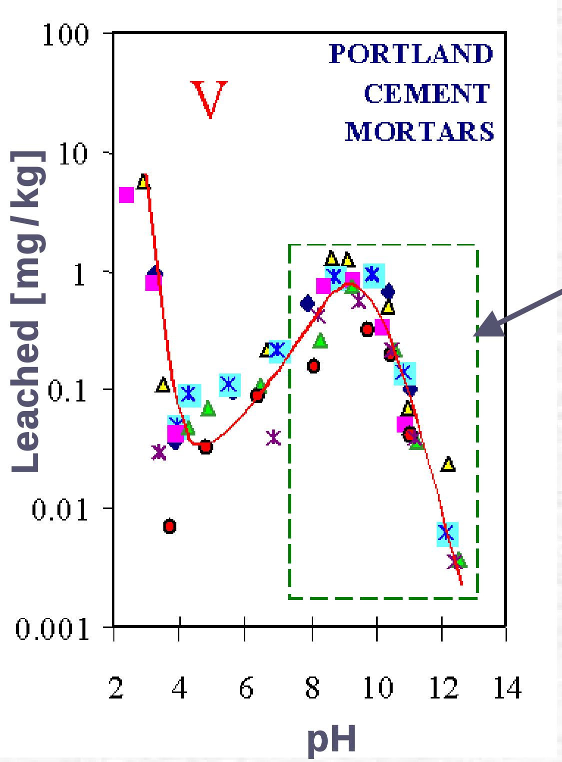




- pH dependence test as reference basis

Consistent Leaching Behavior

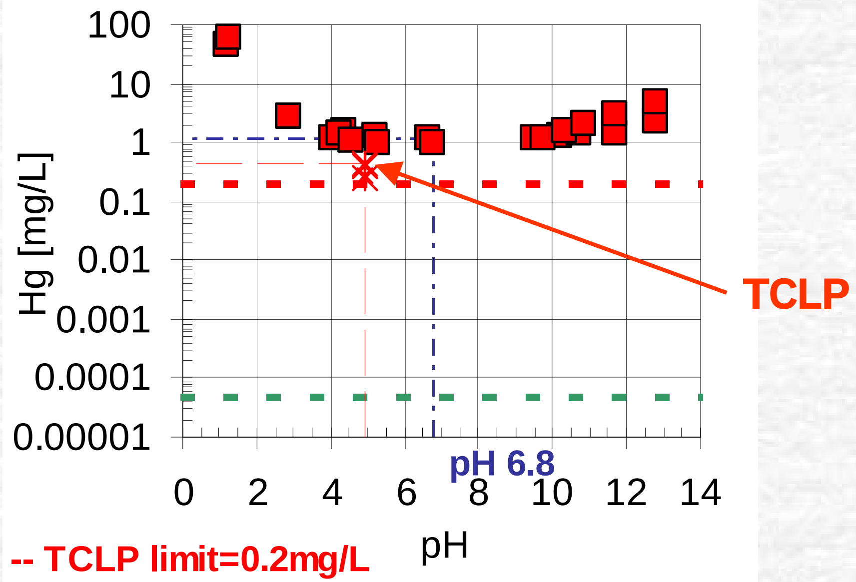


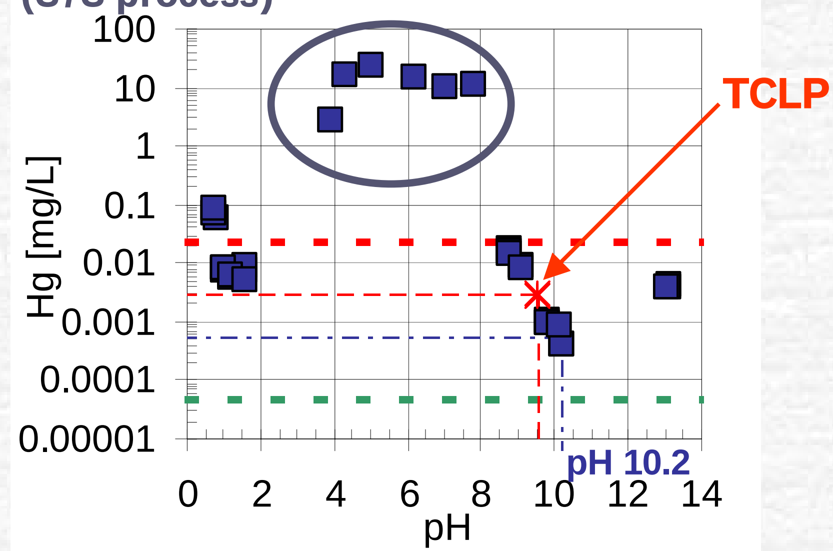


Most relevant pH range for cement-bound materials

A class of materials behaves consistently according to controlling chemistry

Solubility & Release from Untreated and Treated Hg-contaminated Soils





- Vendor 3 treatment resulted in a significant increase in Hg solubility for pH between 4 and 8

Conceptual Approach to Leaching Evaluation

(Kosson, van der Sloot *et al.*, 2002, Environ. Engr. Sci., 19, 159-203)

- Measure intrinsic leaching characteristics of material
- Evaluate release in the context of field scenario
  - External influencing factors such as carbonation, oxidation
  - Hydrology
  - Mineralogical changes
- Use geochemical speciation and mass transfer models to estimate release for alternative scenarios
  - Model complexity to match information needs
  - Many scenarios can be evaluated from single data set

Do NOT mimic field scenarios with specific tests!

Too many tests with limited data comparability!

Measuring Intrinsic Leaching Characteristics

- Aqueous-solid partitioning as a function of pH and Liquid to Solid ratio
  - Batch extractions
  - Constituent fraction readily leached
  - Controlling mechanism for release (mineral dissolution and solubility, solid phase adsorption, aqueous phase complexation)
- Release kinetics
  - Percolation (column tests)
  - Diffusion (monolithic or compacted granular tank leaching tests)
  - Use results in conjunction with understanding of pore water chemistry to determine mass transfer rate constants (e.g., effective diffusivities)

Main Types of Leaching Tests

- Equilibrium-based leaching tests
  - Carried out on size reduced material
  - Aim to measure contaminant release related to specific chemical conditions (pH, LS ratio)
- Mass transfer-based leaching tests
  - Carried out either on monolithic material or compacted granular material
  - Aim to determine contaminant release rates by accounting for both chemical and physical properties of the material
- Percolation (column) leaching tests
  - May be either equilibrium or mass transfer rate

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Equilibrium Characterization

Solubility and Release as a Function of pH (SR002.1)

- 11 parallel solubility extractions
- DI with HNO_3 or KOH addition
- Size reduced material
- Contact time based on size
- LS ratio: 10 mL/g dry
- Endpoint pH
  - Distributed 3≤pH≤12

Particle size	Contact time
< 0.3 mm	18 hr
< 2 mm	48 hr
< 5 mm	168 hr

Titration curve and constituent solubility or release curves

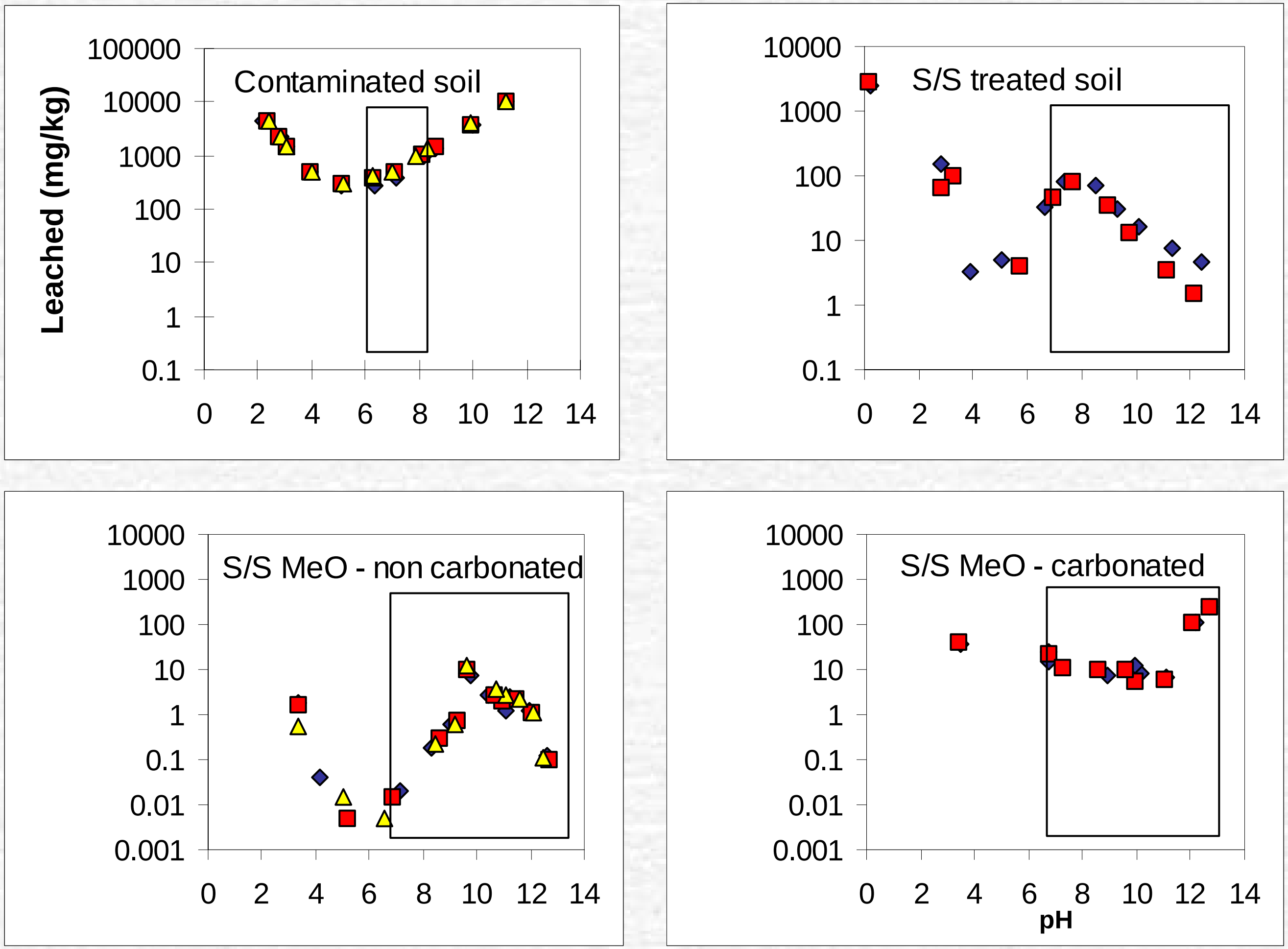
Equilibrium Characterization
Solubility and Release as a Function of LS (SR003.1)

- 5 parallel extractions
- DI water
- Size reduced material
- Contact time based on particle size
- LS ratios
  - 0.5, 1, 2, 5, and 10 mL/g dry

Particle size	Contact time
< 0.3 mm	18 hr
< 2 mm	48 hr
< 5 mm	168 hr

Estimate of constituent concentration in the pore water

Arsenic Leaching Behavior From Soils and Solidified Wastes



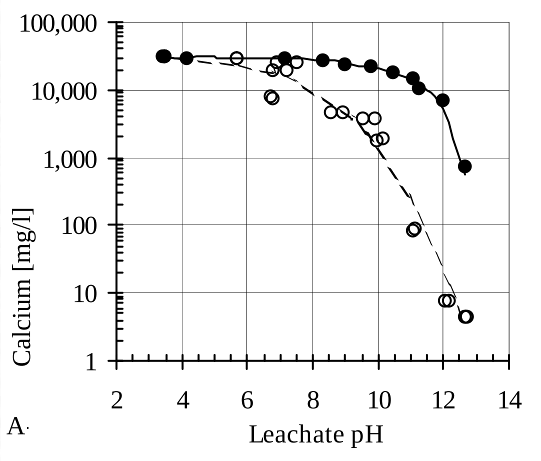
Same soil untreated & treated

Same S/S treated metal Oxide matrix with & Without exposure to Carbon dioxide (carbonation)

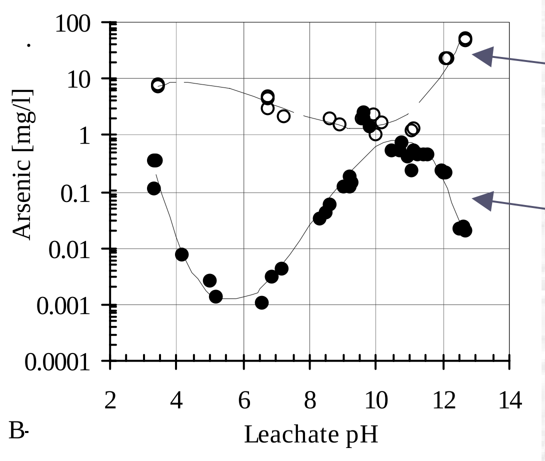
Carbonation and Matrix Solubility

Portland Cement Matrix

Ca



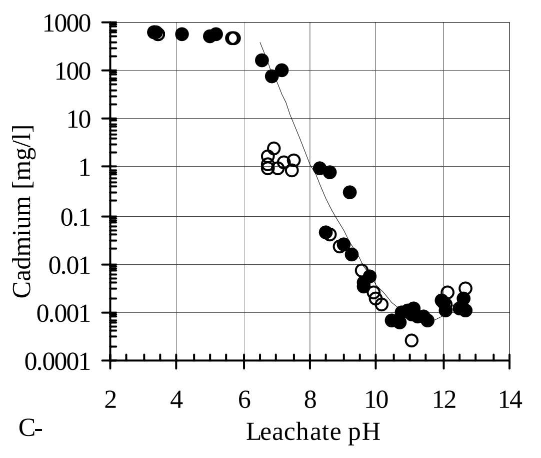
As



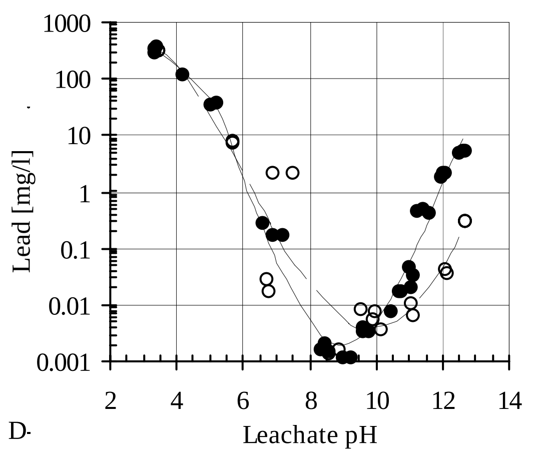
Carbonated

Without carbonation

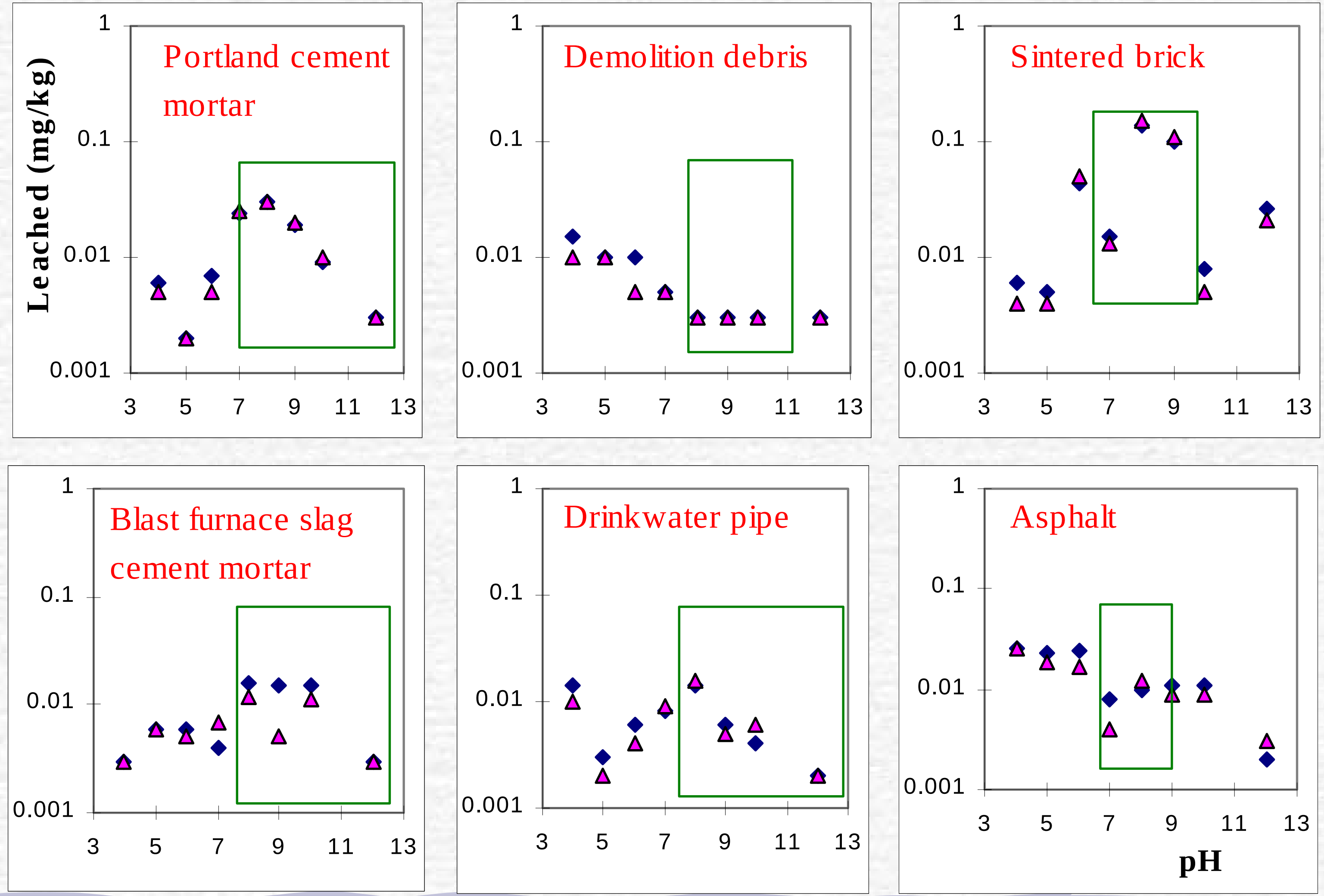
Cd



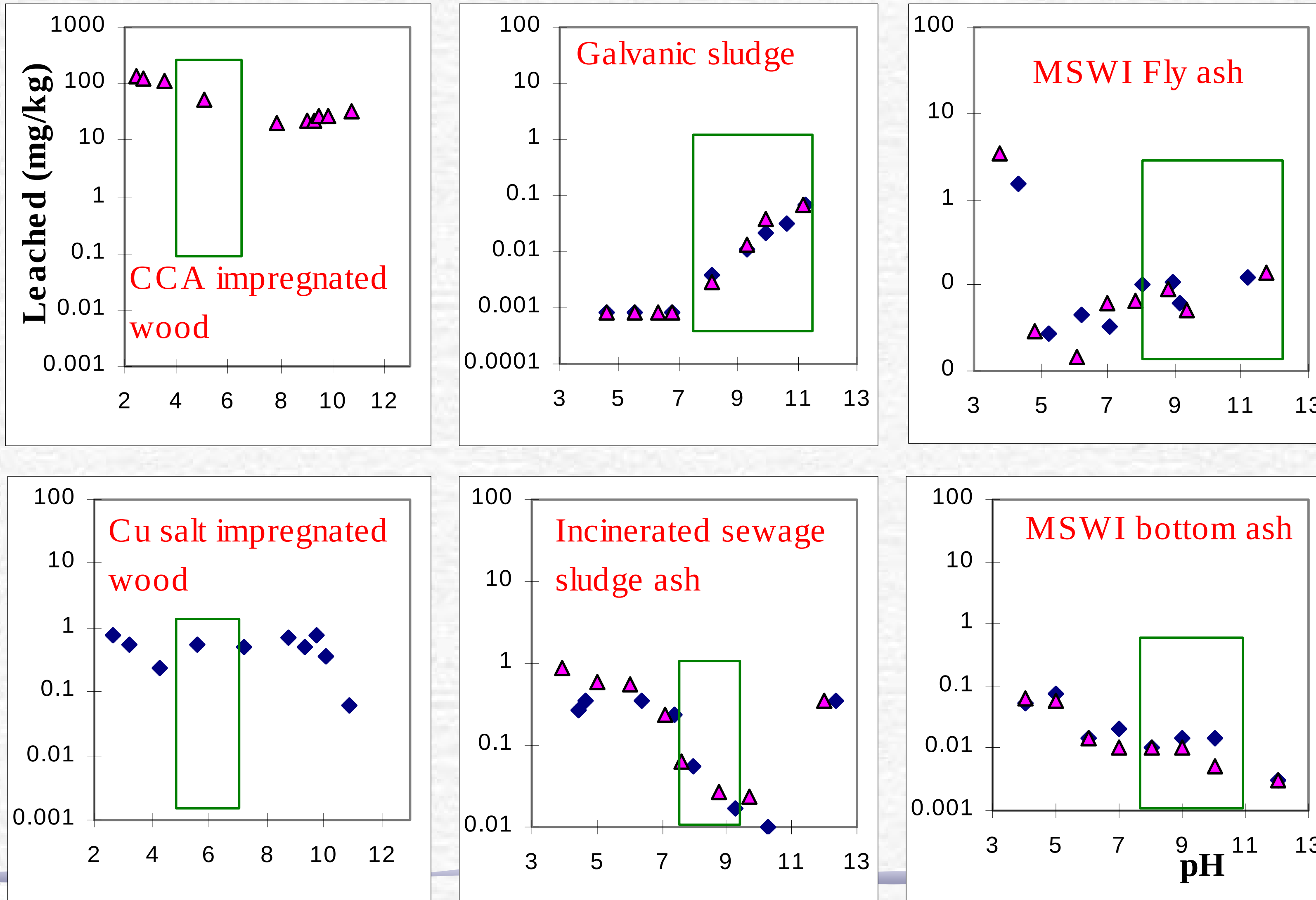
Pb



Arsenic Leaching Behavior From a Range of Construction Materials



Arsenic Leaching Behavior From Treated Wood and a Few Waste Types



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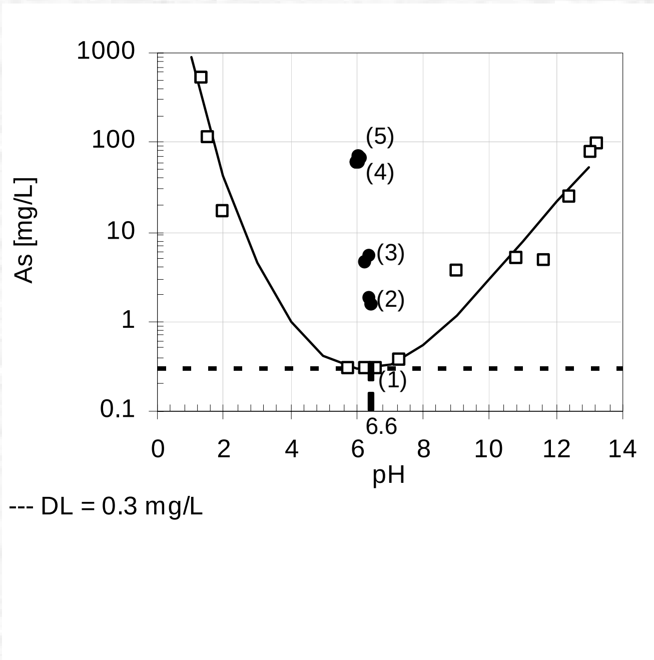
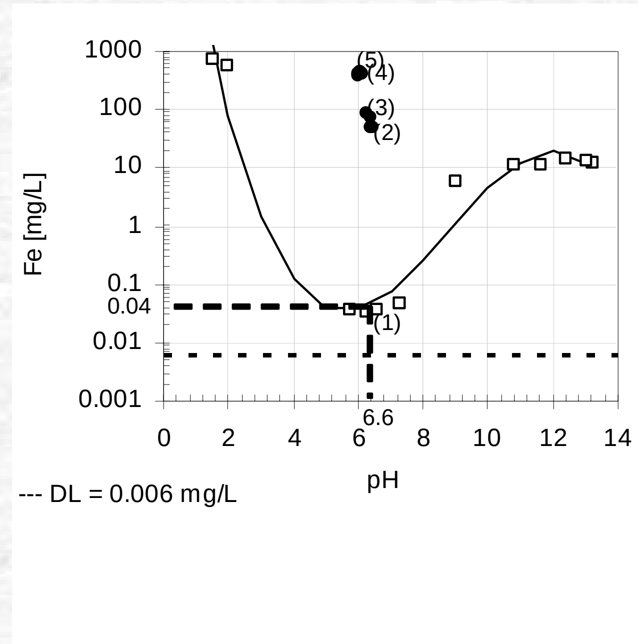
Use of Sodium Ascorbate as a Reducing Agent (Mining Waste)

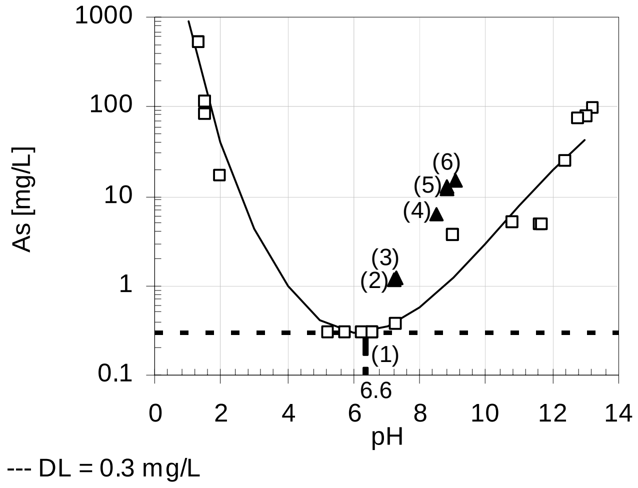


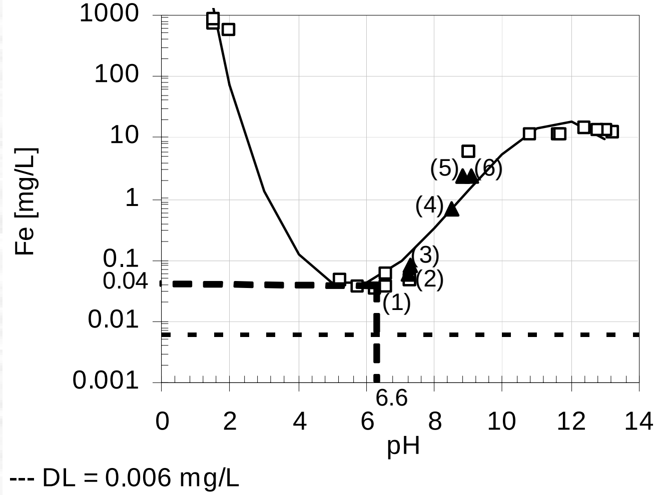


(1) Baseline, ORP = 410mV vs. NHE
(2) 0.0075 mol L^{-1}, ORP = 340 vs. NHE
(3) 0.01 mol L^{-1}, ORP = 345mV vs. NHE
(4) 0.025 mol L^{-1}, ORP = 5mV vs. NHE
(5) 0.046 mol L^{-1}, ORP = -7mV vs. NHE

Chatain and Sanchez, 2005

Use of Sodium Borohydride as a Reducing Agent (Mining Waste)



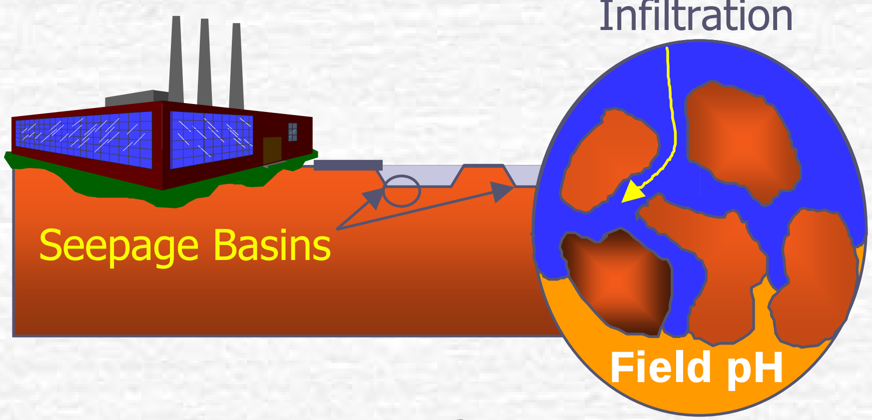


(1) Baseline, ORP = 410 mV vs. NHE
(2) 0.0075 mol L-1, ORP = 140 mV vs. NHE
(3) 0.01 mol L-1, ORP = 150 mV vs. NHE
(4) 0.025 mol L-1, ORP = -440 mV vs. NHE
(5) 0.046 mol L-1, ORP = -445 mV vs. NHE
(6) 0.075 mol L-1, ORP = -500 mV vs. NHE

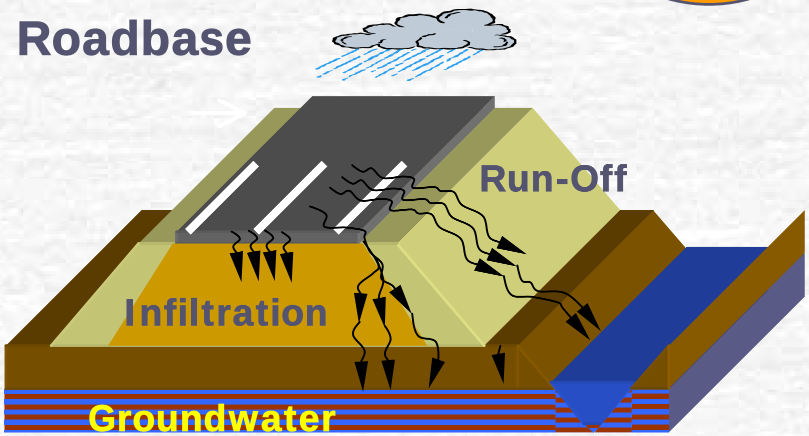
Chatain and Sanchez, 2005

Release Modes

Contaminated soil



Roadbase

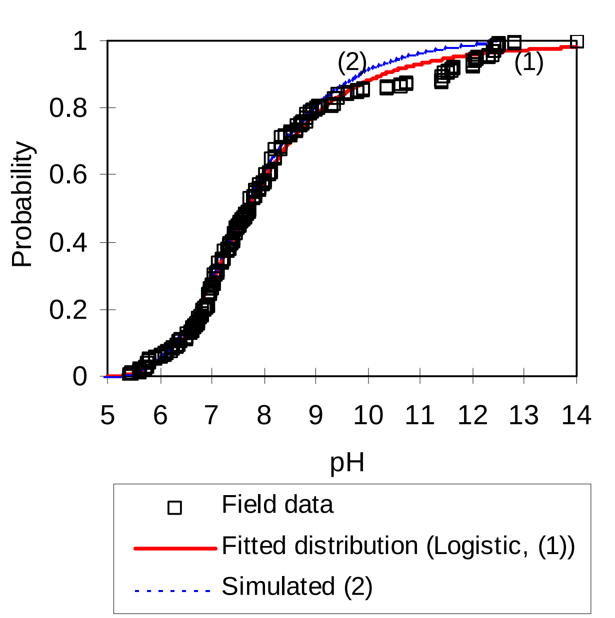


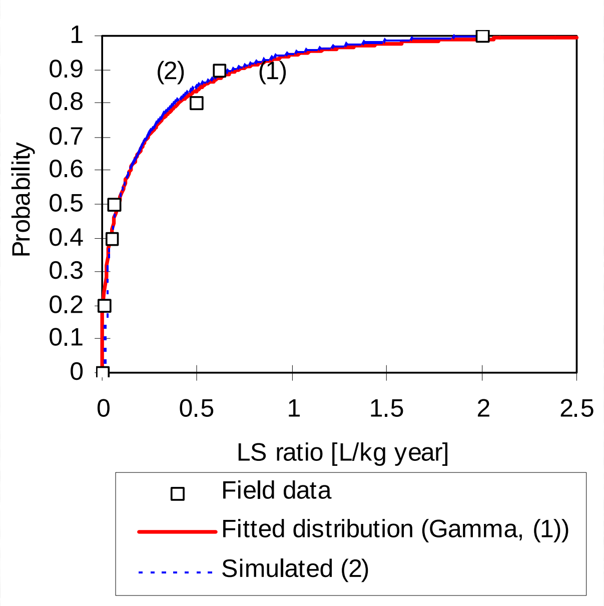
- **Percolation through granular materials**
  - Granular or highly permeable material
  - Local equilibrium controls release
  - Preferential flow may be important
- **Flow around low permeability (monolithic) materials**
  - Coupled diffusion and pore-water chemistry controls release
  - Boundary conditions are important

Long-term Assessment Models

- Simple release models
  - Percolation/equilibrium model
  - Diffusion model
- More sophisticated release models to account for
  - Chemistry between solid-liquid phases (empirical or geochemical speciation)
  - Effect of intermittent wetting
  - Effect of external stresses (e.g., carbonation)
- Use of probabilistic approach to allow for
  - Consideration of a range of management sites and conditions
  - Consideration of a range of expected climate conditions and waste characteristics
  - Bounded levels of confidence and distribution frequencies for release estimates

Probabilistic Assessment of Release for Land Disposal (based on USEPA database)





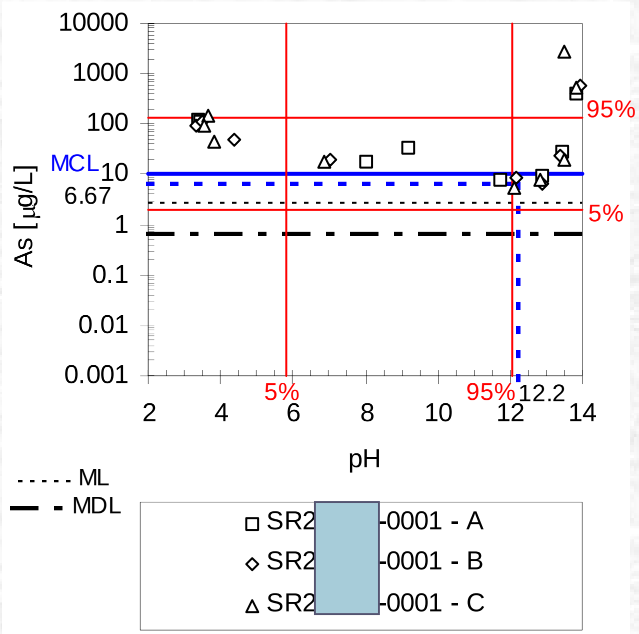
pH	Field data	Fitted distribution	Simulated
pH min	5.40	4.74	4.92
pH - 5%	5.80	6.00	5.97
pH - 95%	12.09	11.62	10.63
Mean pH	8.10	8.13	7.86
pH max	12.80	+infinity	12.50

LS 1 year	Field data	Fitted distribution	Simulated
LS min	1.0E-05	3.3E-04	3.3E-04
LS - 5%	5.5E-04	4.9E-04	4.4E-04
LS - 50%	0.06	0.08	0.08
LS - 95%	1.50	1.07	1
LS max	2.50	+infinity	1.99

Example
Arsenic Leaching as a Function of pH

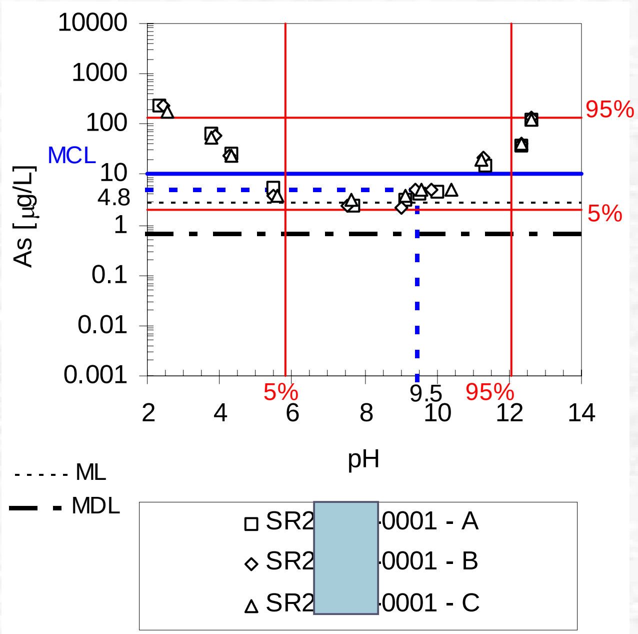
Baseline

As total content*: 80.5 ± 1.9 μg/g

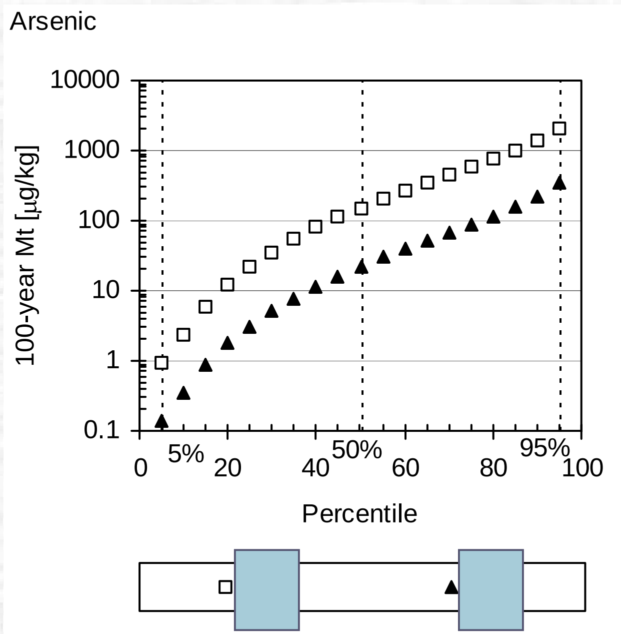


w/ activated carbon injection

As total content*: 27.9 ± 2.1 μg/g



Example
Probability Distribution of 100 yr Release Estimates for Land Disposal



	PB		PT	
	µg/kg	%	µg/kg	%
Mt min	0.2	0.0003	0.1	0.0003
Mt - 5%	0.9	0.0011	0.1	0.0005
Mt - 50%	152	0.2	22	0.0772
Mt - 95%	2095	2.6	338	1.2
Mean Mt	468	0.6	90	0.3
Mt max	4693	5.8	10157	36.4

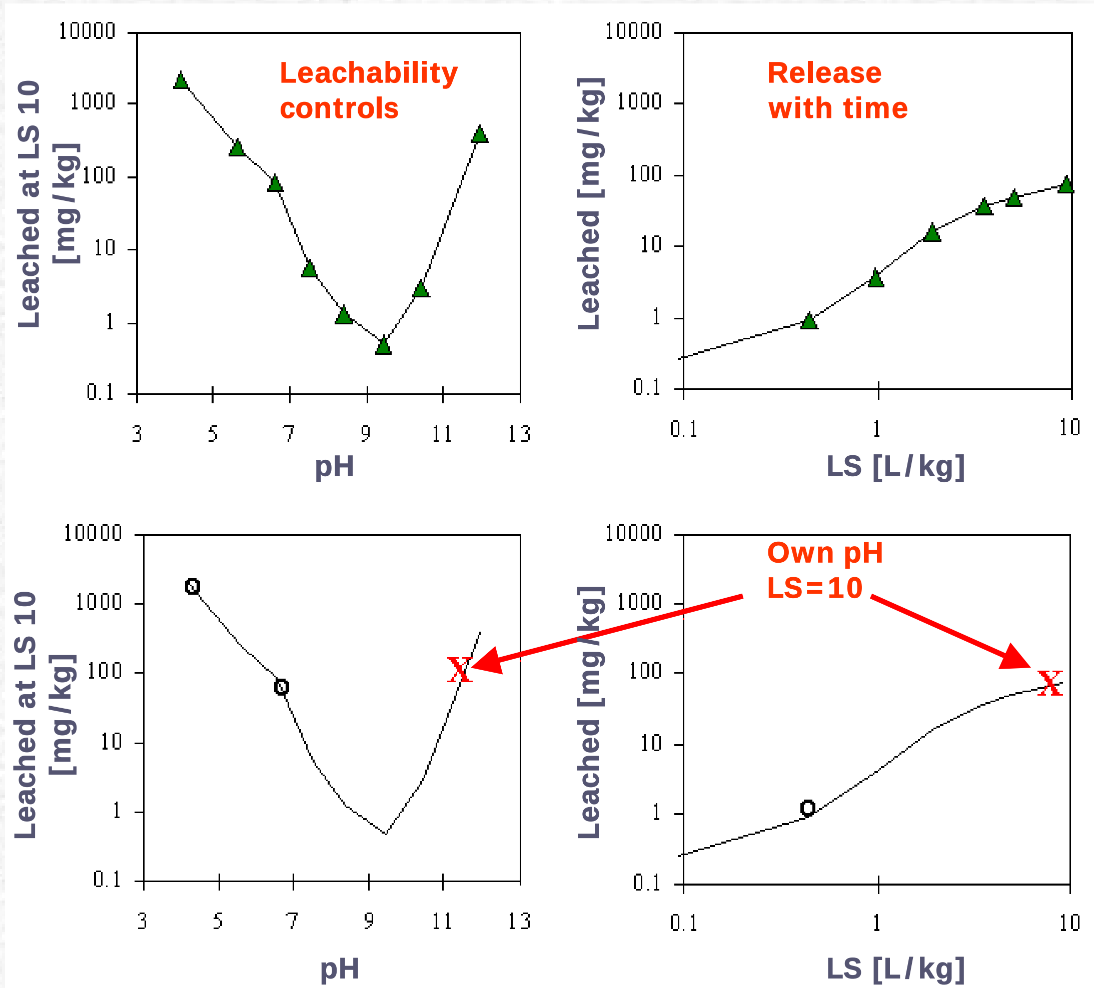
Hierarchy in Testing

- In the context of detailed characterization of a material, use simplified testing for compliance and quality control purposes
  - Establish material performance criteria for specified application
  - Test material to establish
    - Performance consistent with initial characterization (new material source or significant process change?)
    - Performance consistent with management acceptance criteria
  - Limit testing to critical parameters
  - Establish material quality control monitoring program

Hierarchy in Testing
Characterization and Compliance Tests

Characterization

Compliance



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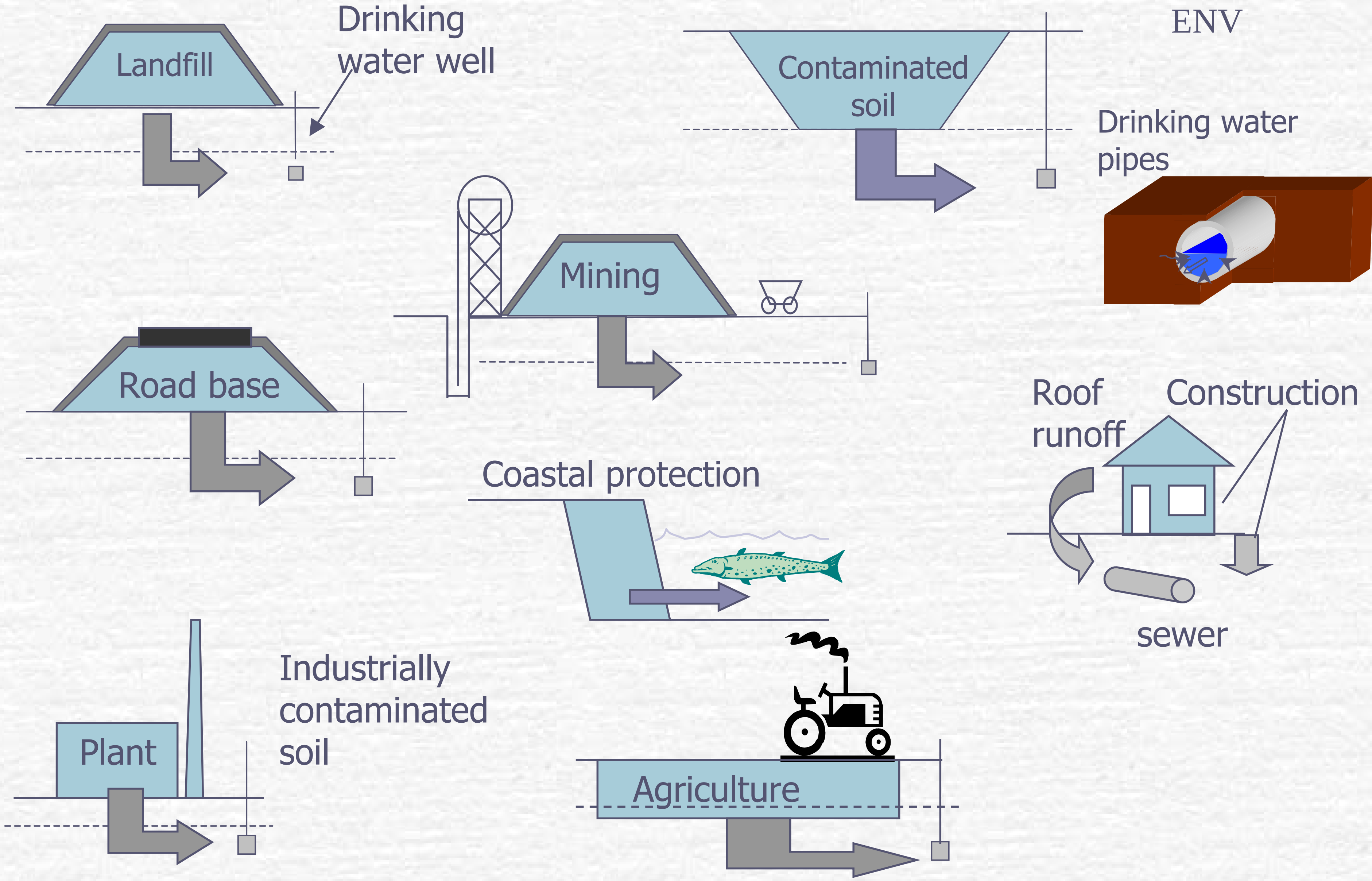
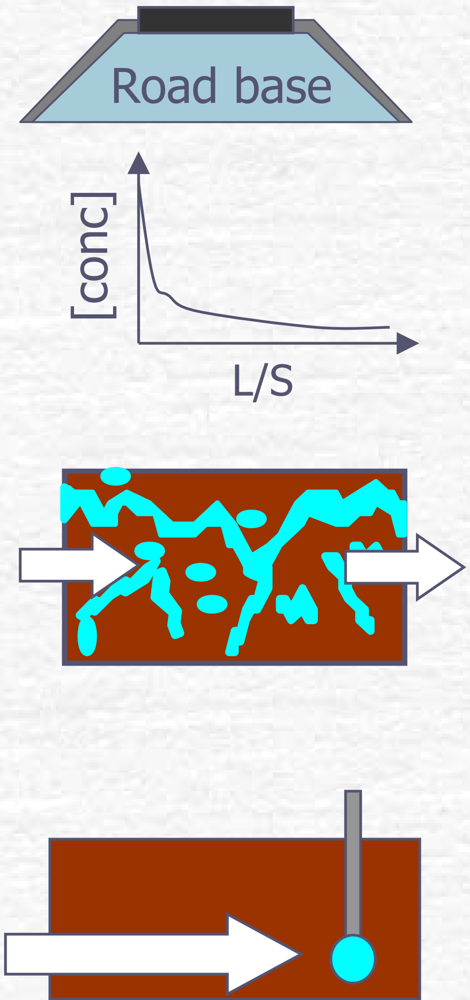
DIFFERENT IMPACT SCENARIOS……..

16 Dec. 2003 DG ENV



.... SIMILAR PROBLEM



SOURCE TERM

↓

TRANSPORT

↓

IMPACT

Different for each scenario, material, changes over time (carbonation, redox), etc.

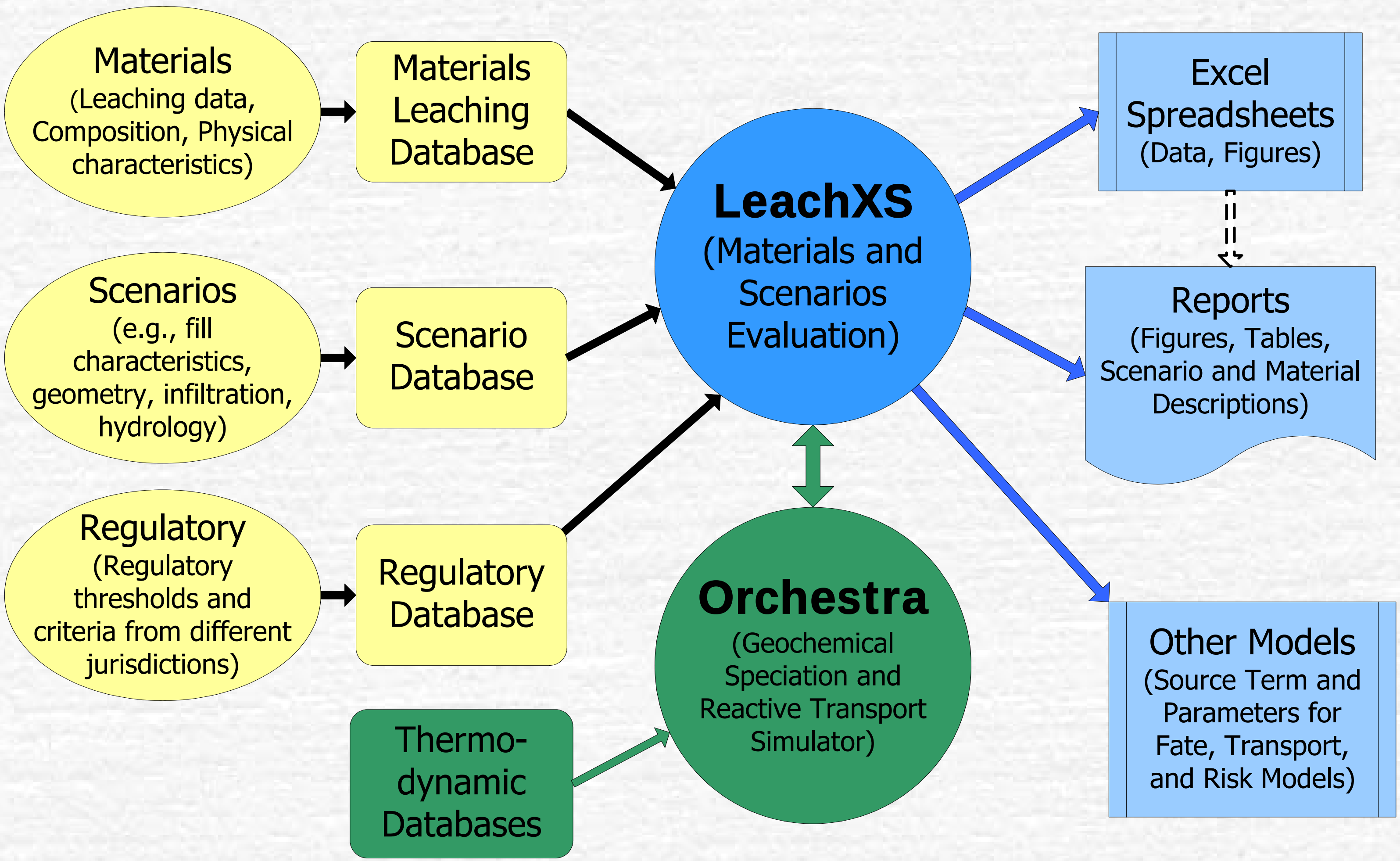
Transport in unsaturated zone and saturated zone to point of compliance similar for each scenario

Point of compliance

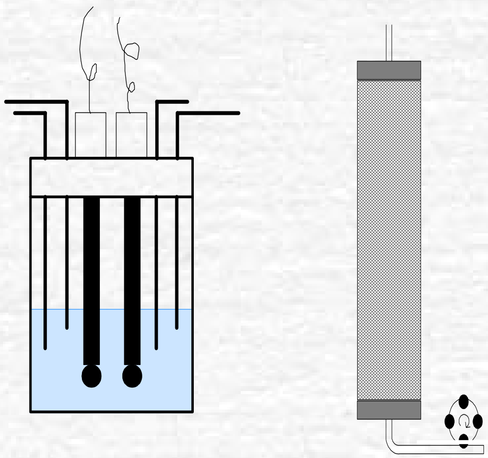
LeachXS[1] as a Decision Support Tool to Manage and Interpret Test Data

[1]**LeachXS** is a software tool being jointly developed by The Netherlands Energy Research Foundation (ECN), Vanderbilt University (USA) and DHI Water and Environment (Denmark), including test methods selection, database, data presentation, evaluation, geochemical speciation modeling, and scenario assessment.

LeachXS Structure



GEOCHEMICAL MODELING

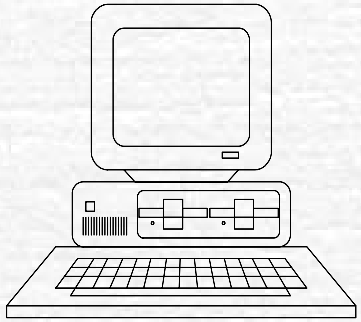


pH stat test

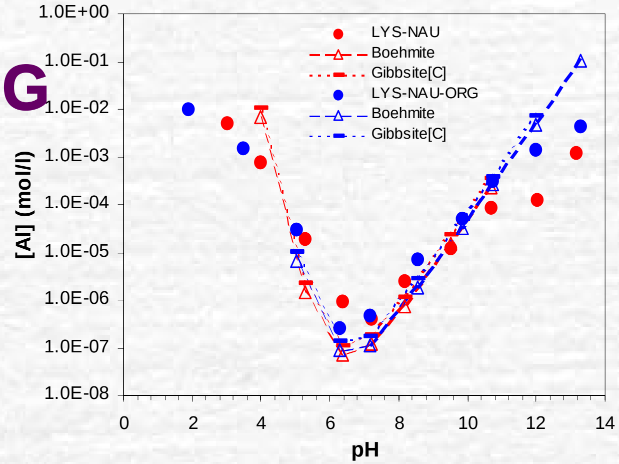
L/S = 10, t = 48 hr

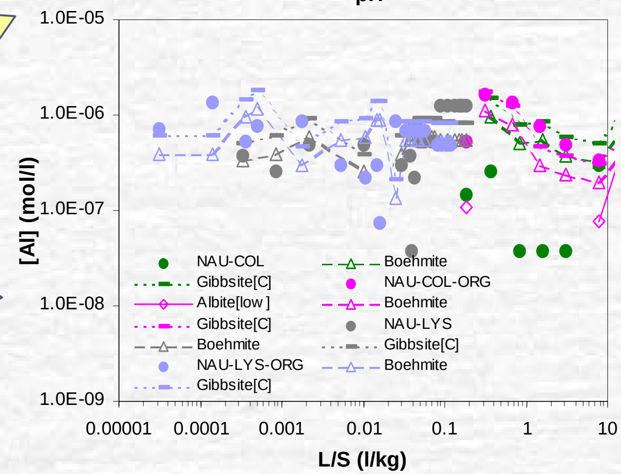
Percolation test
L/S=0.1-10

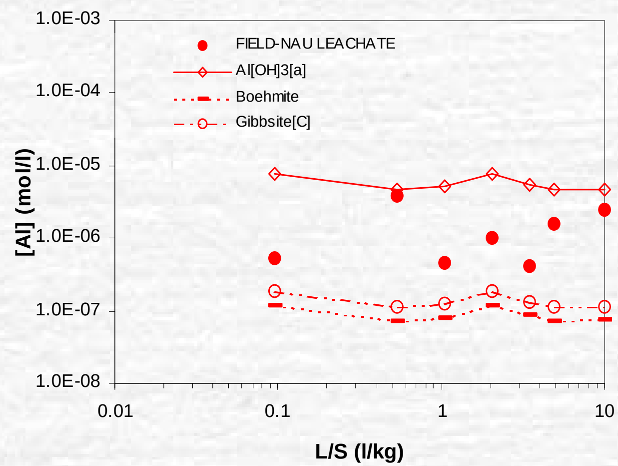
Field leachate



Geochemical modeling



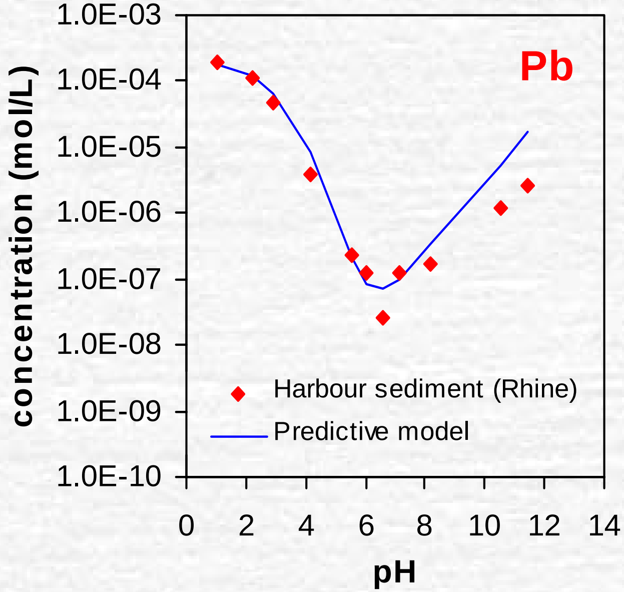


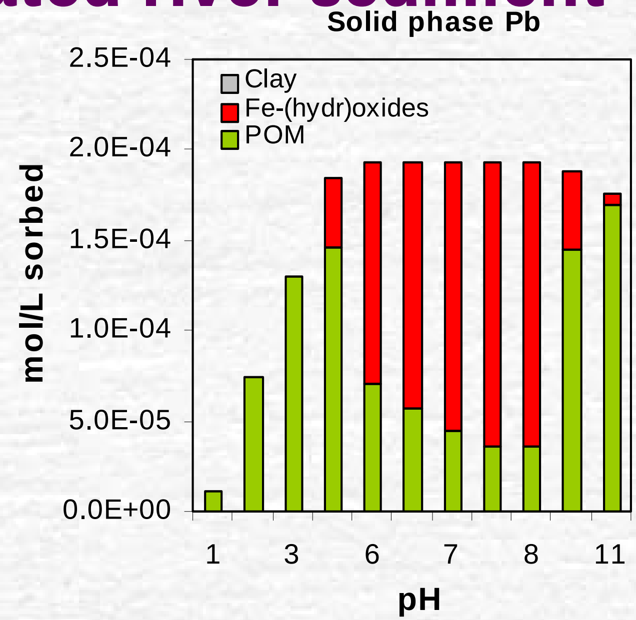


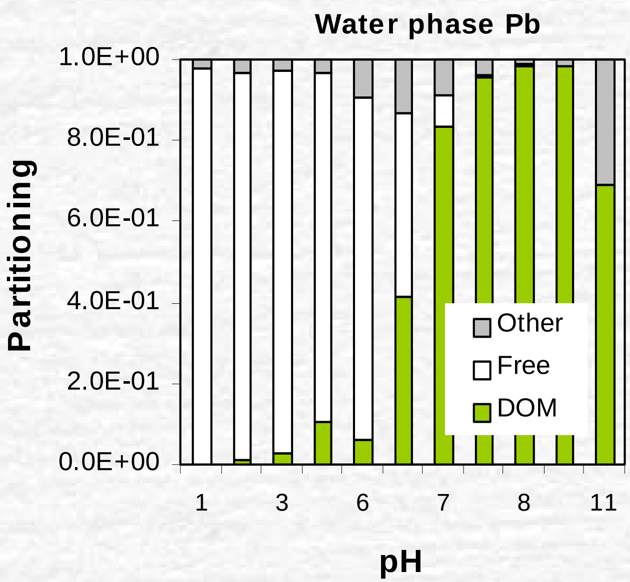
ORCHESTRA with extended MINTEQ database + Nicca Donnan

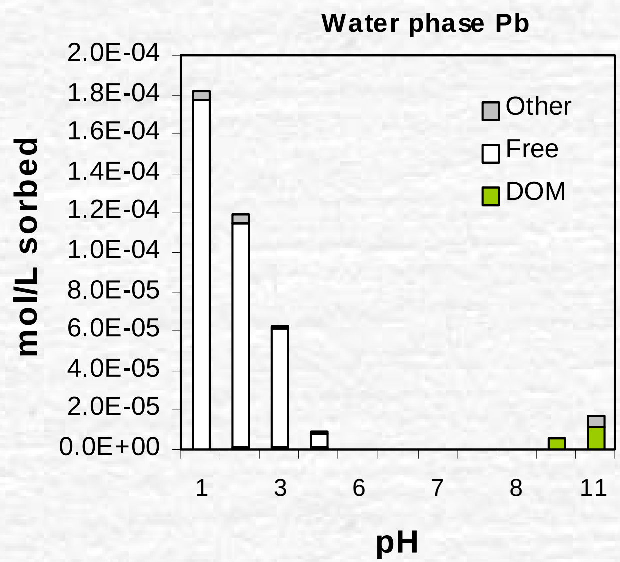
LeachXS

Modelled solid and liquid phase speciation of Pb in a contaminated river sediment









SEDNET Workshop
June 10, 2004
San Sebastian, Spain

LeachXS

USEPA Evaluation of Coal Combustion Residues from Facilities with Enhanced Mercury Control

- Extensive QAQC program development
  - Methods validation with mass balance on reference CCR
  - QAQC within leaching methods, chemical analysis, data evaluation
- Leaching characterization
  - Release as function of pH (SR002.1)
  - Release as function of LS (SR003.1)
  - Mass transfer release (when considered necessary)
- Comparison with field data
  - Leachate concentrations reported in USEPA database
  - Field sampling from CCR management facilities
- Release Scenario Assessment
  - Land disposal
  - Probabilistic release estimates based on range of conditions (pH, LS) reported in USEPA database (improved estimates to be based on EPRI data)
  - Release estimates for default scenarios at 3 pHs (acid, alkali, own)

Key Messages

- Measure intrinsic leaching characteristics, use geochemical speciation and mass transfer models in conjunction with management scenarios to estimate constituent release.
- Use results to assess impacts, develop acceptance criteria and monitoring strategies.
- The tools exist and data are currently being obtained to achieve assessments for specific scenarios of residue disposal. Tailoring for specific uses is needed.
- "Classes" of drinking water treatment residues likely can be identified. After initial characterization, simplified testing (i.e., an index test) can be used to confirm conformance with previously defined class.

Acknowledgements

- USEPA
  - Office of Solid Waste
  - National Risk Management Research Laboratory (RTP)
  - Northeast Hazardous Research Center
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- EU Research Program
  - Network Harmonization of Leaching and Extraction Tests
- Netherlands Ministry of Housing, Spatial Planning and Environment (VROM)