Environmental Dynamics Modeling and Cesium Removal Techniques

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Why Modeling?

- Monitoring
  - Provides the current contamination level
  - Data accepted by the public
  - Does not predict future contamination levels, especially under significant changes on climate, land, and water channel conditions
  - Provides calibration and validation data for modeling
  - Time consuming and expensive
- Modeling
  - Predicts future contamination levels
    - Under expected weather and other normal conditions
    - Under hypothetical, especially extreme conditions, e.g.,
      - Extreme storms/floods and drought conditions
      - Accident conditions
  - Selects effective remediation actions
  - Public is skeptical to modeling
    - Need to convince public on modeling
    - Reproduce past monitoring results through model calibration and validation
    - Present model results in easily understandable forms (e.g., video)
  - Modelers need to understand phenomena they are trying to simulate
  - Modelers need to know what and how models predicts

Three-Dimensional Surface Water Contaminant Transport Code

FLESCOT code

- Time-varying, three-dimensional code to simulate

In water column

- Velocity and water depth affected by river flow, tide, wind, waves, water temperature, and salinity
- Turbulent kinetic energy and its dissipation
- Water temperature
- Salinity
- Transport, deposition, and re-suspension of each of sand, silt and clay, separately
- Dissolved contaminant (radionuclides, pesticides, heavy metals, toxic chemicals and aqueous chemical species) with interactions with sand, silt and clay: (adsorption/desorption)
- Transport, deposition, and re-suspension of particulate contaminants adsorbed by each of sand, silt and clay

Within bed of river, estuary, lake, sea, and ocean

- 3-d distributions of river, estuary, lake and sea bed elevation changes due to sediment deposition and re-suspension (bed erosion)
- 3-d distributions of sand, silt and clay fractions within the bed
- 3-d distributions of contaminants each associated with sand, silt, clay within bed

One- and Two-Dimensional Surface Water Codes

TODAM and FETRA codes

- Time-varying, one-dimensional and two-dimensional codes to simulate

  In water column:

  - Velocity and water depth affected by river flow and tide; (TODAM only)
  - Transport, deposition, and re-suspension of each of sand, silt and clay, separately
  - Dissolved contaminant (radionuclides, pesticides, heavy metals, toxic chemicals) with interactions with sand, silt and clay: (adsorption/desorption)
  - Transport, deposition, and re-suspension of particulate contaminants adsorbed by each of sand, silt and clay

  Within the bed of river, estuary and sea:

  - 2- and 3-d distributions of river, estuary, lake and sea bed elevation changes due to sediment deposition and re-suspension (bed erosion)
  - 2- and 3-d distributions of sand, silt and clay fractions within the bed
  - 2- and 3-d distributions of contaminants each associated with sand, silt clay within the bed

Examples of Surface Water Transport Modeling with These Codes

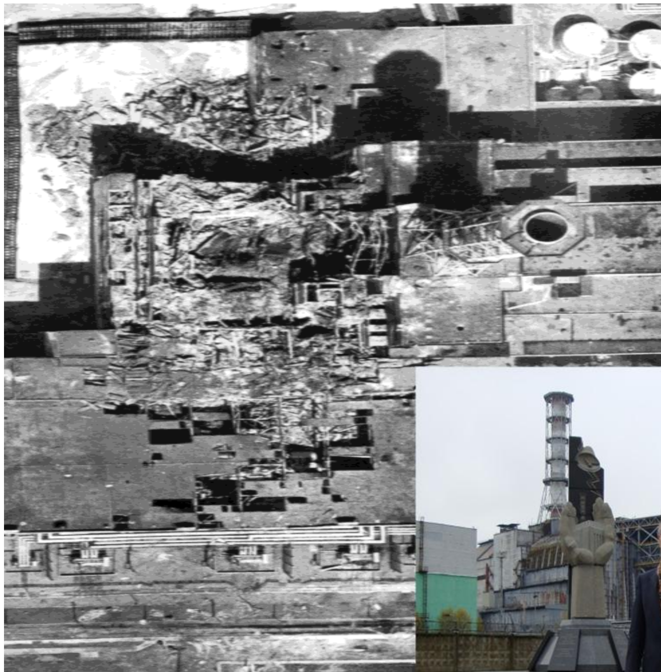
- Large Rivers
  - Columbia River in WA (^{65}Zn)
  - Tennessee and Clinch Rivers, TN (^{137}Cs)
  - Pripyat and Dnieper Rivers in Ukraine for the Chernobyl Accident Assessment (^{137}Cs, ^{90}Sr, $^{238, 239, 240}$Pu, ^{241}Am)
  - Ob, Irtysh, Tobal, Iset and Techa Rivers in Russia (^{137}Cs, ^{90}Sr, $^{238, 239, 240}$Pu)
- Medium Rivers
  - Ukedo River and its tributaries in Fukushima (^{137}Cs)
  - Cattaraugus, Buttermilk, and Frank Creeks in NY (^{137}Cs, ^{90}Sr)
  - Yazoo, Big Sunflower, Tallahatchie, and Coldwater Rivers in MS (pesticide, Toxaphere)
- Small Rivers
  - Mortandad and Los Alamos Canyons in NM (^{239}Pu)
  - Four Mile and Wolf Creeks in IA (pesticide, Alachlor)
  - Monticello Stream Channels in MN (toxic chemical, Dioxin)
- Reservoirs and Lakes in Fukushima
  - Kido Dam Reservoir (^{137}Cs)
  - Ogi Dam Reservoir (^{137}Cs)
  - Ogaki Dam Reservoir (^{137}Cs)

Examples of Surface Water Transport Modeling with These Codes

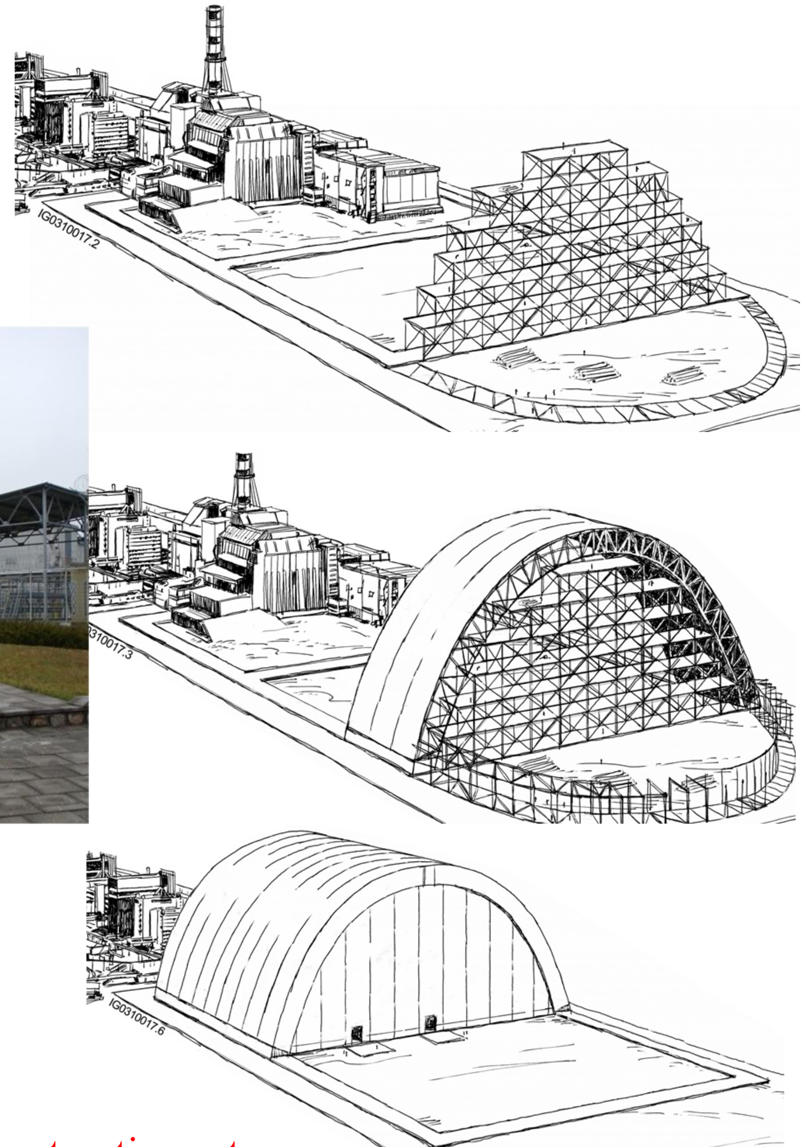
- Coastal Water, Seas, and Oceans
  - Pacific Coast of Japan ($^{238, 239}Pu$, ^{137}Cs)
  - Irish Sea (^{137}Cs)
  - Kara Sea, Russia (^{137}Cs, ^{90}Sr)
  - 2,800-m deep Radionuclide Disposal Site in Atlantic Ocean off NY (^{137}Cs)
  - Buzzard Bay and New Bedford Harbor in MA (PCB, heavy metals)
  - Strait of Juan De Fuca and Sequim Bay in WA (waste water)
  - Beauport Sea, AK (temperature, salinity)
  - 4000-m Deep Pacific Ocean (Disposed CO_2)
  - South Florida Offshore and Near-shore Waters in FL (spilled oil)
  - San Diego Bay in CA (heated water)
- Estuaries
  - Hudson River Estuary in NY (^{137}Cs)
  - James River Estuary in VA (pesticide, Kepone)

Chernobyl Nuclear Plant and its New Safe Confinement

At Accident

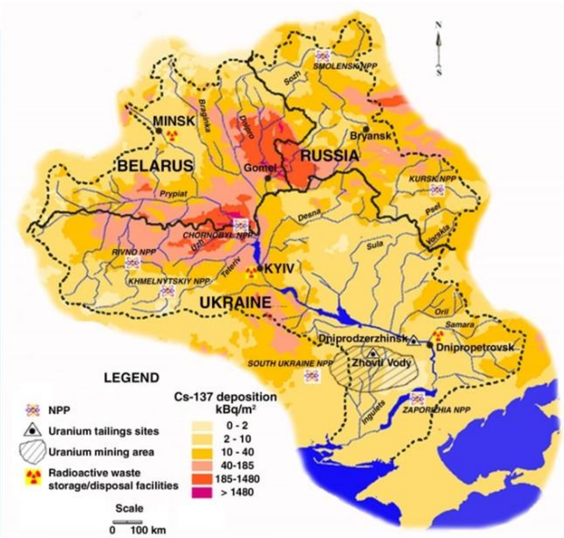


Current



NSC construction set-up

Purpose: Assess the Dnieper River Contamination Level Dnieper and Pripyat River Modeling with TODAM Code



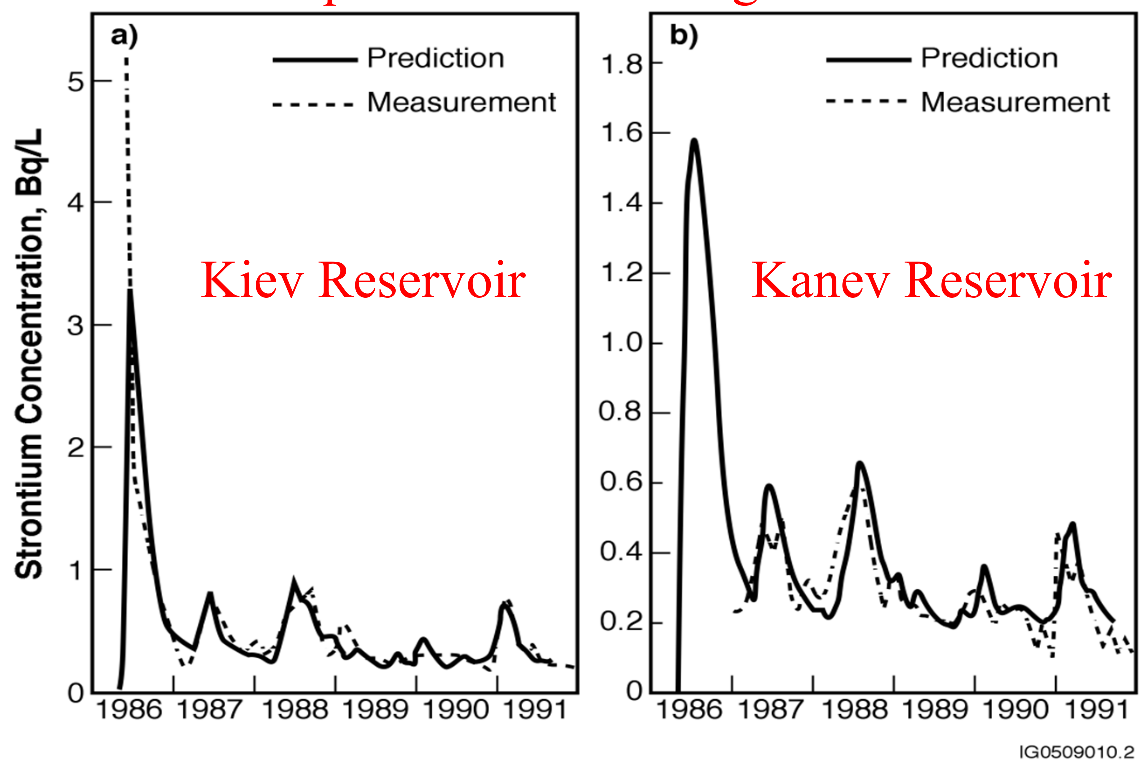
The Dnieper River water affects 20 million Ukrainians:

- Drinking water
- Irrigation water

It is critical to evaluate the Dnieper River contamination

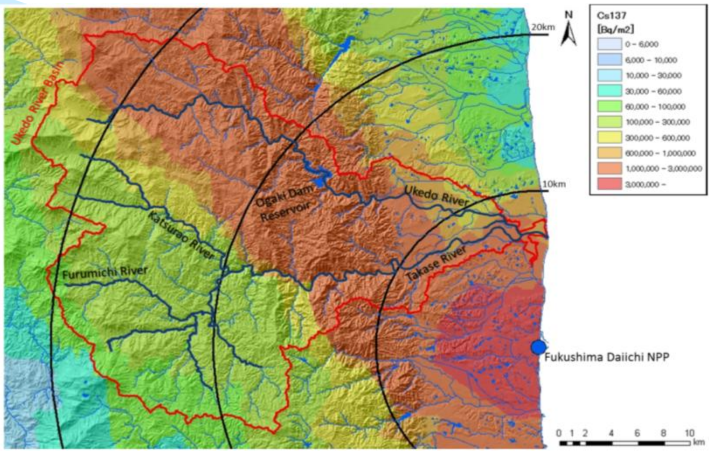
TODAM was validated with Chernobyl data

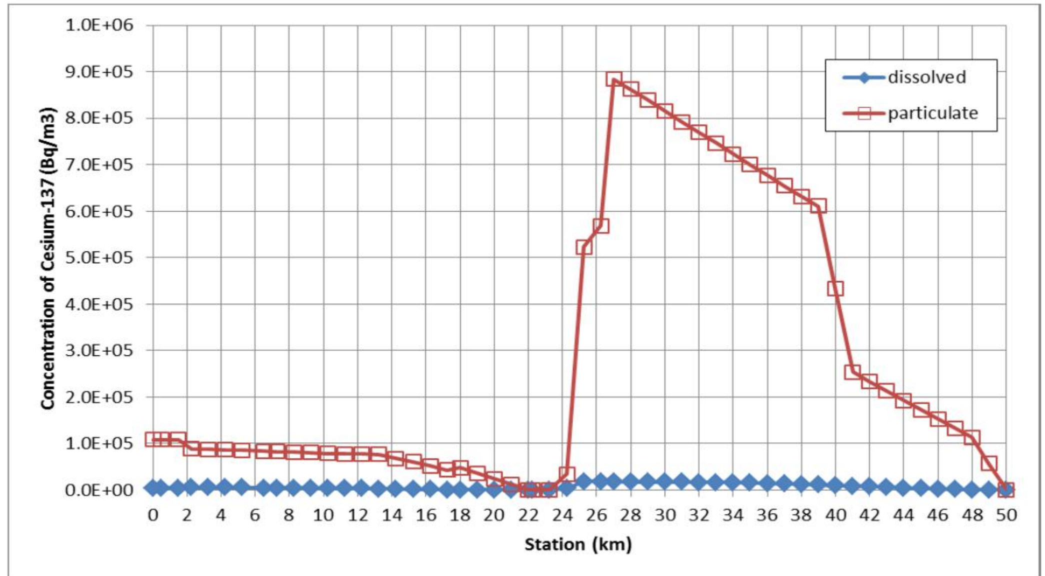
Chernobyl ^{90}Sr Modeling
Importance of flooding and reservoirs

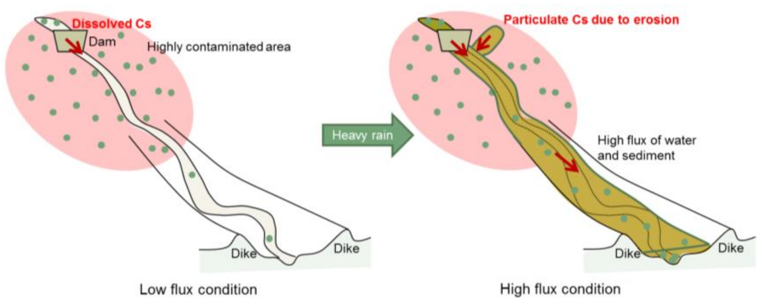


Purpose: Evaluate ^{137}Cs Movement in the Ukedo and Takase Rivers with TODAM Model:
The importance of high flows, sediment transport and the reservoir

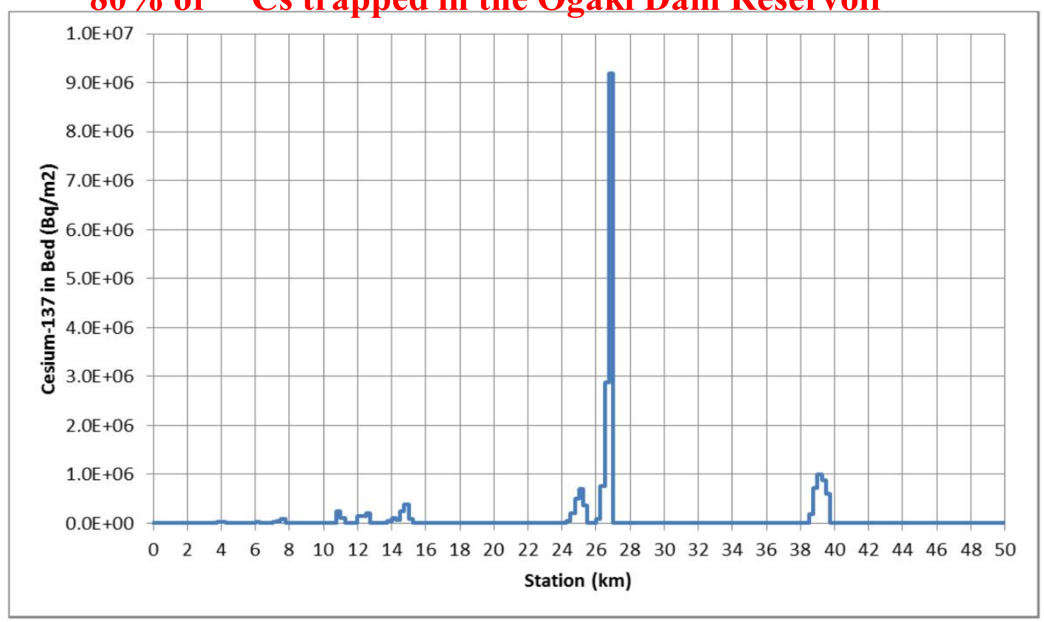
Joint Study with JAEA

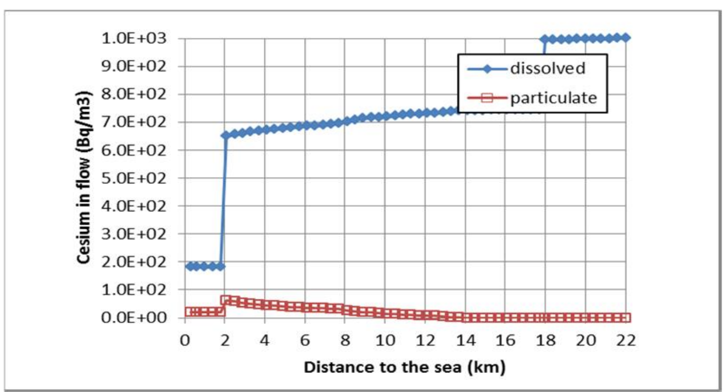






80% of ^{137}Cs trapped in the Ogaki Dam Reservoir



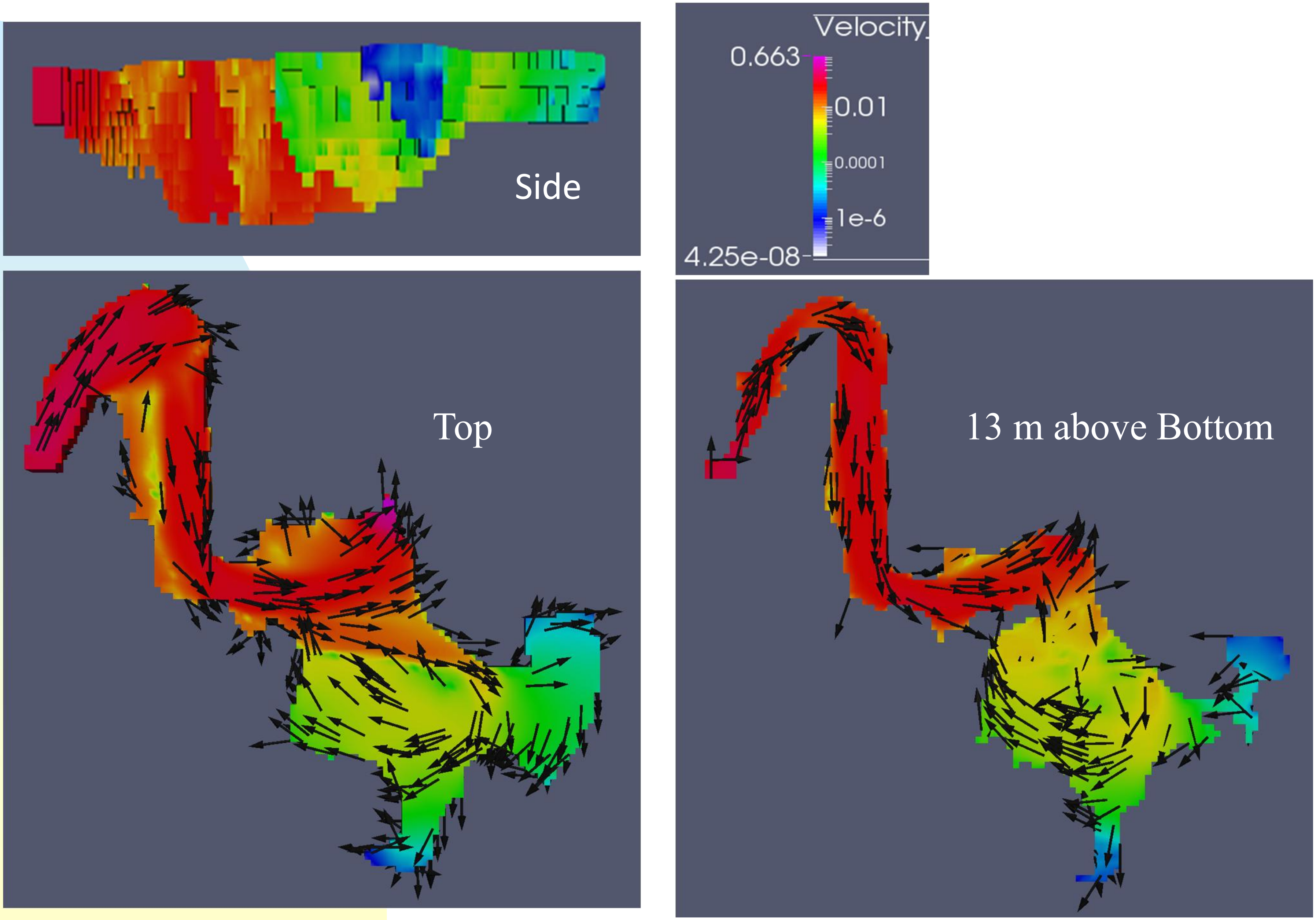


Ogaki Dam Reservoir: Critically Important Water Resource

- The Ogaki Dam Reservoir provides **water to rice paddies** through numerous irrigation canals and ditches
- Thus, it is very important to keep reservoir **water clean**
- Some irrigation water **returns to other rivers (e.g., Odaka River)** that are not as contaminated as the Ukedo River
- It is important **not to spread** ^{137}Cs from the Ogaki Dam Reservoir to areas not contaminated as much.

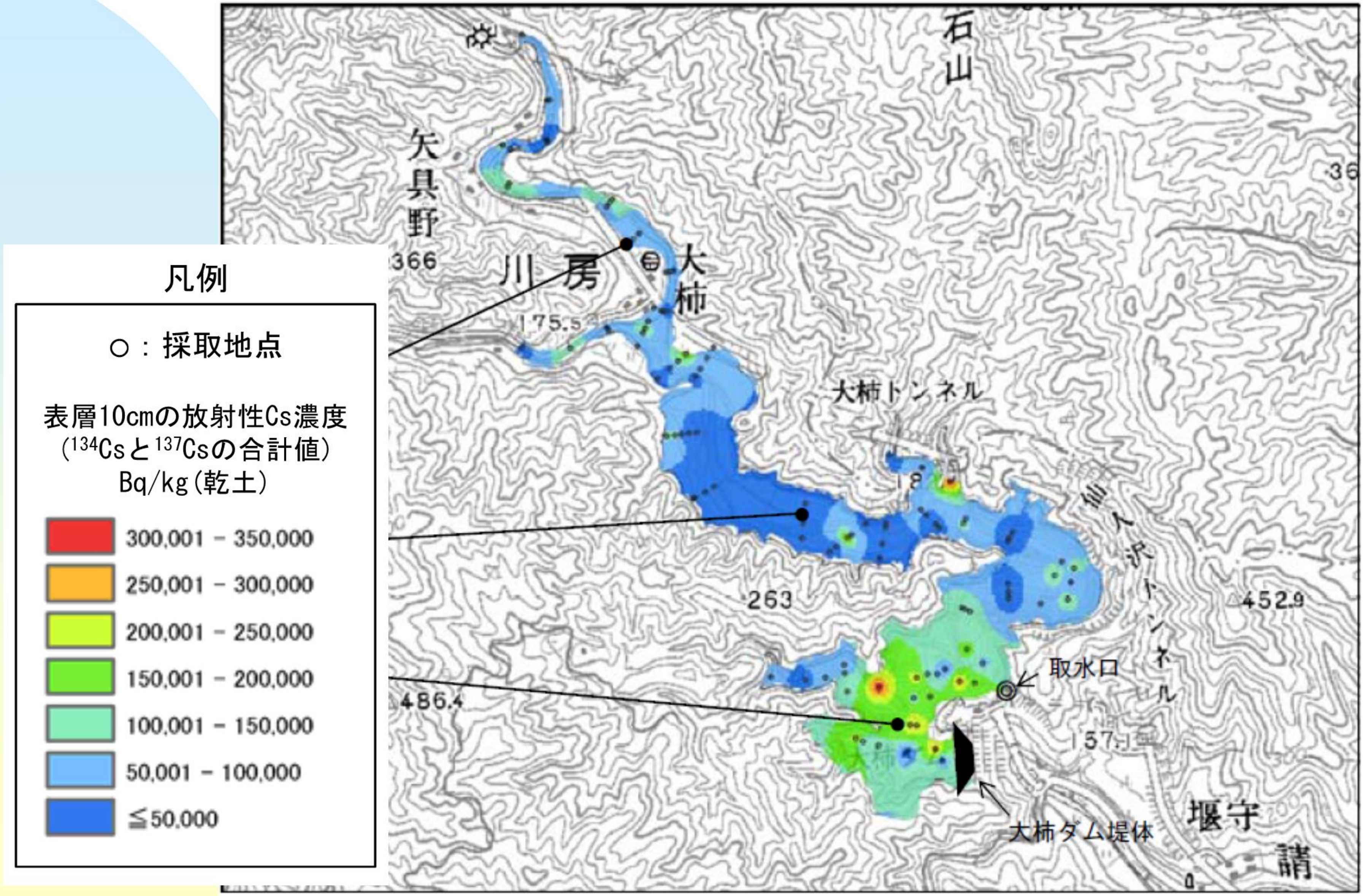
Purpose:Closely Examine Ogaki Dam Reservoir with 3-D FLESCOT Code

Joint Study with JAEA

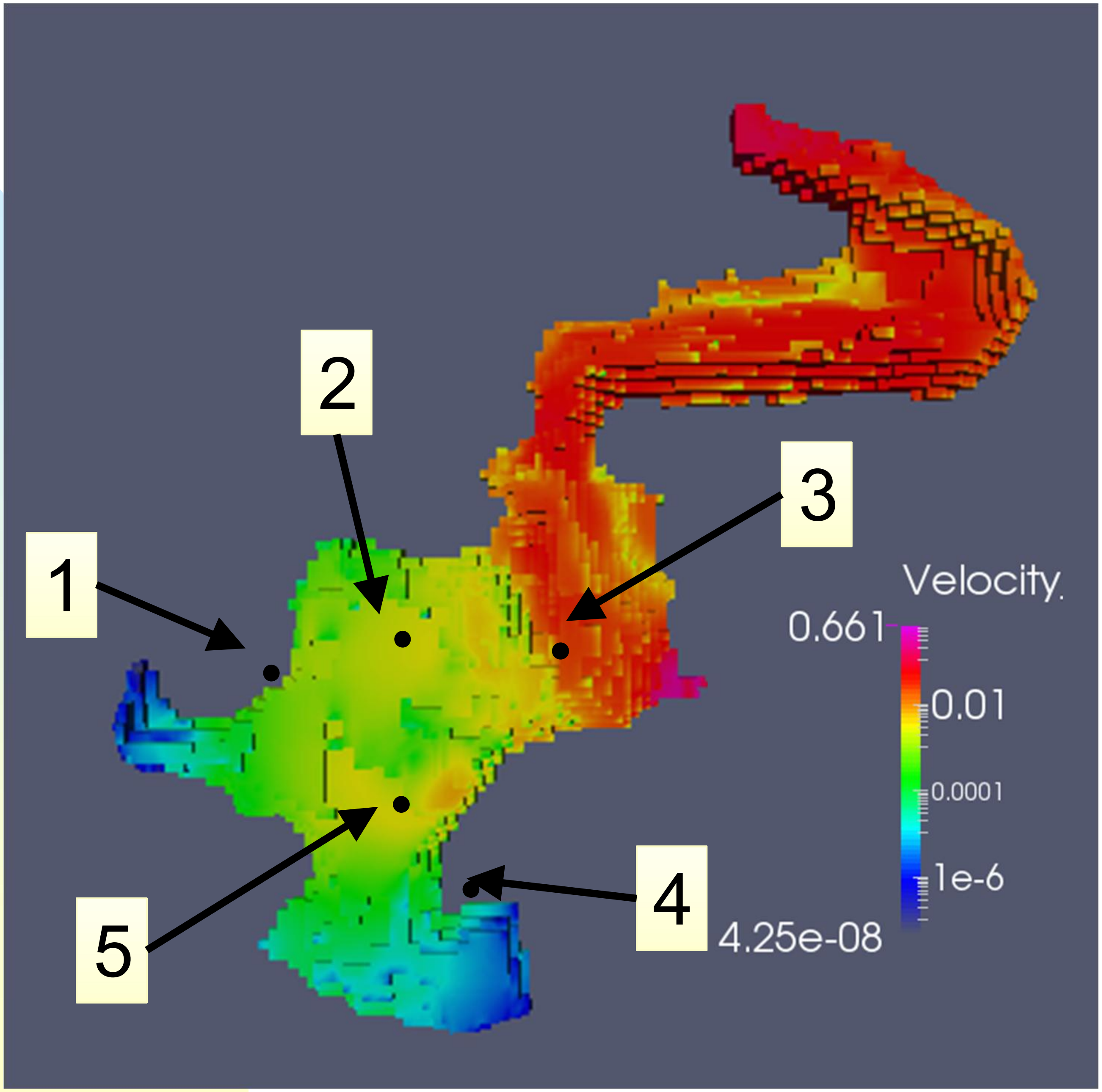


Locations with High Cesium Concentrations in Ogaki Dam Reservoir Bottom

(Data Provided by JAEA)

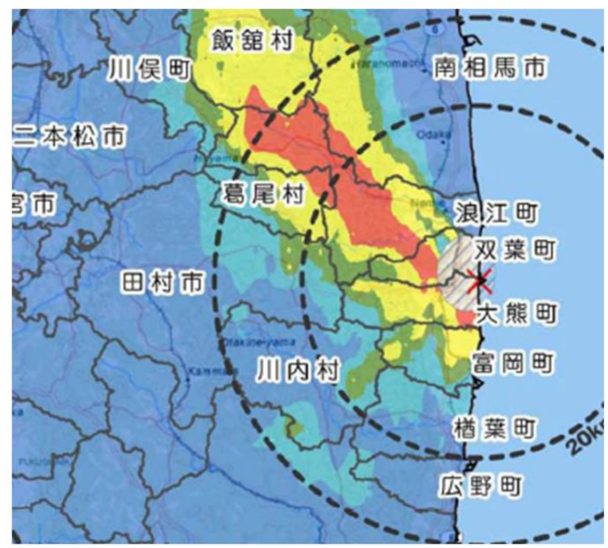


Predicted near (13-m above) Bottom Velocity

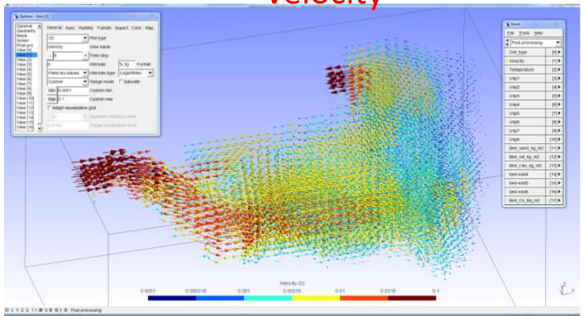


Purpose: Evaluate Cesium Movement in Ogi Dam Reservoir

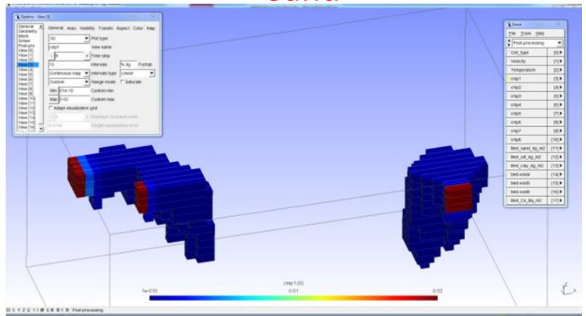
Joint Study with JAEA



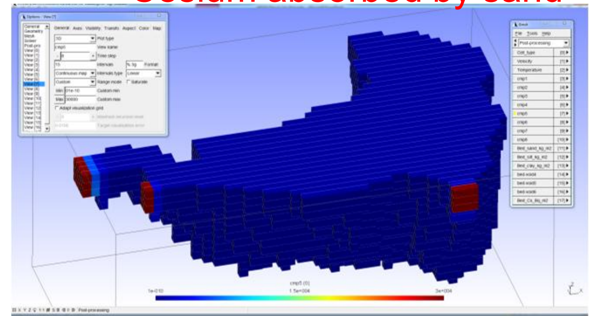
Velocity



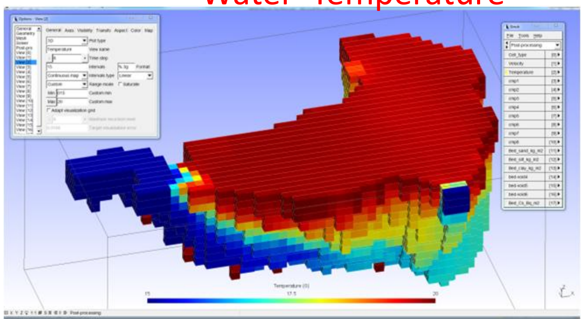
Sand



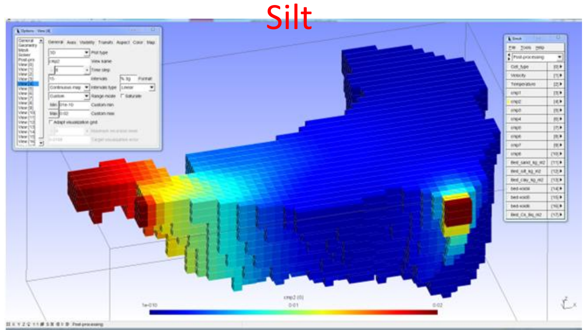
Cesium absorbed by sand



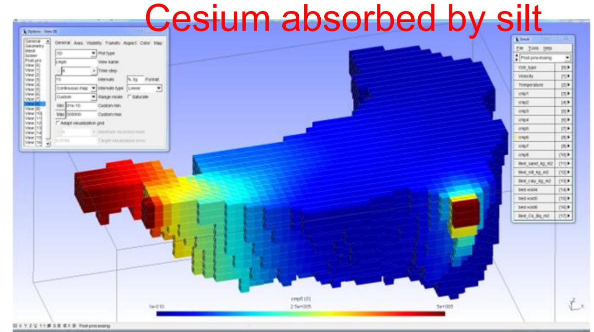
Water Temperature



Silt



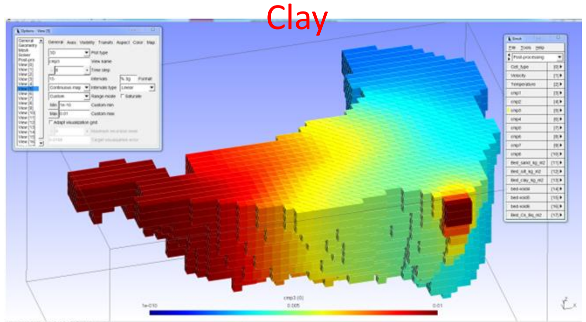
Cesium absorbed by silt



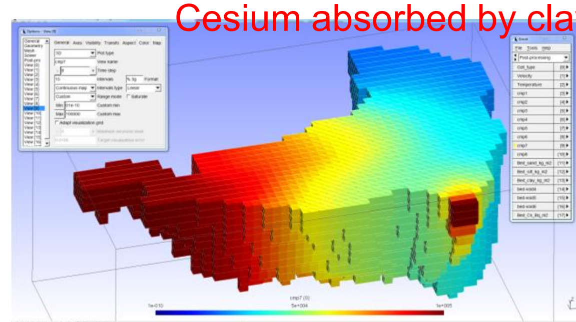
Predicted Cesium Movement

Colder river water gets into warmer reservoir water

Clay

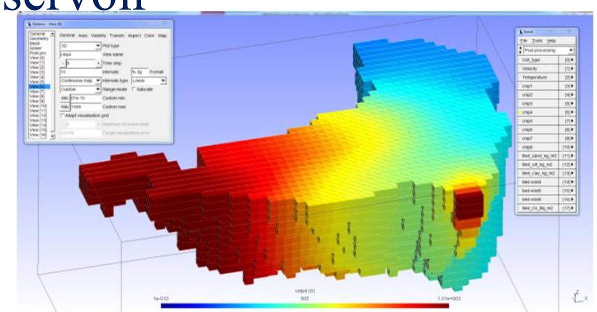


Cesium absorbed by clay



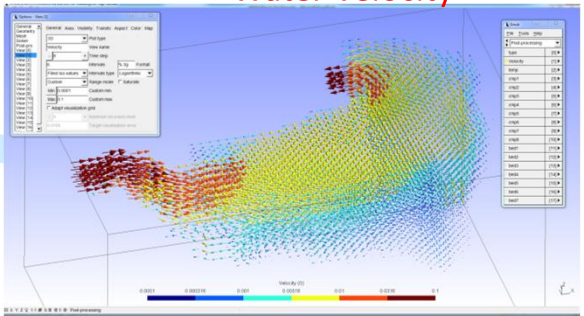
Cesium adsorbed by sand and silt are deposited in the reservoir

Dissolved cesium

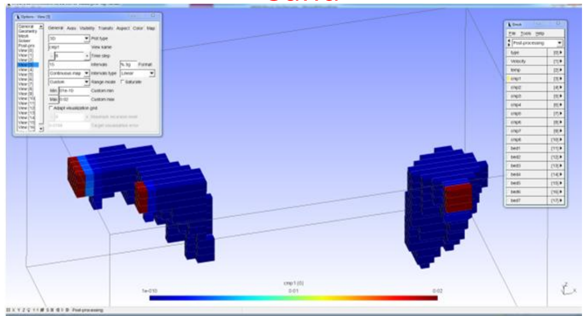


Simulation of the Ogi Dam Reservoir with 3-D FLESCOT

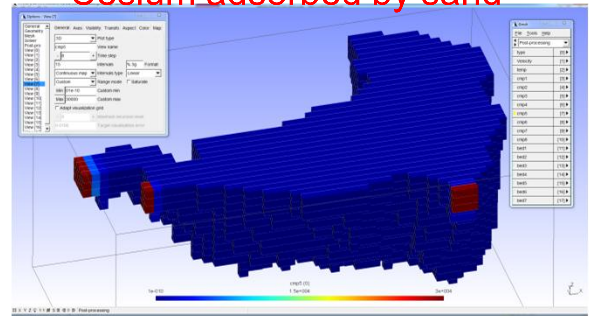
Water Velocity



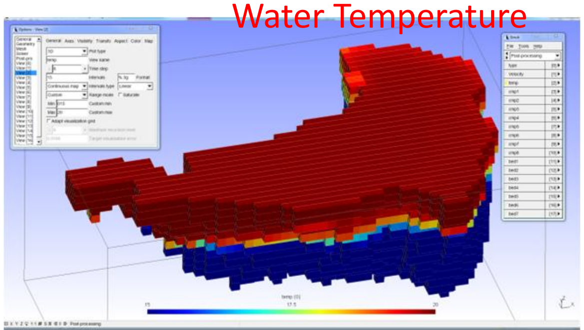
Sand



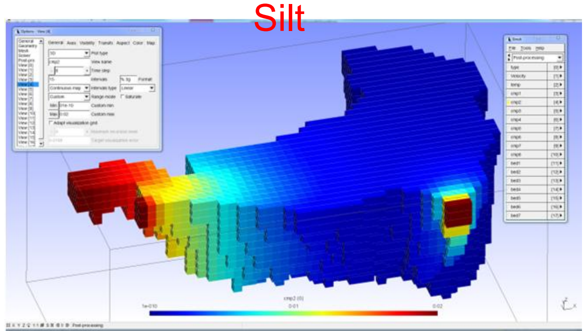
Cesium adsorbed by sand



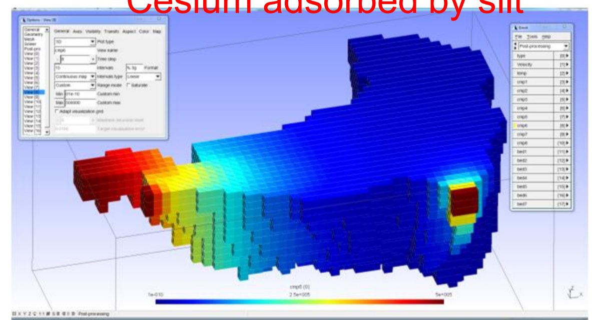
Water Temperature



Silt



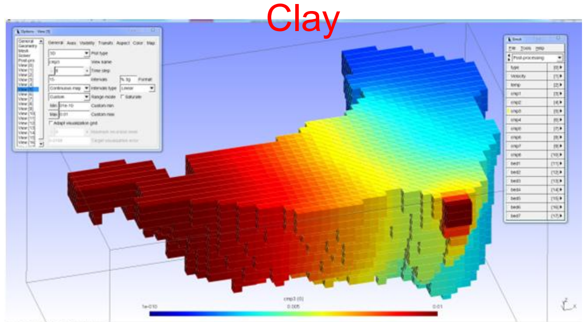
Cesium adsorbed by silt



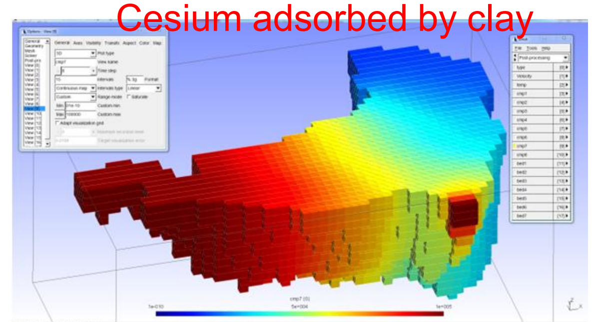
Predicted Cesium Movement

Warmer river water gets into colder reservoir water

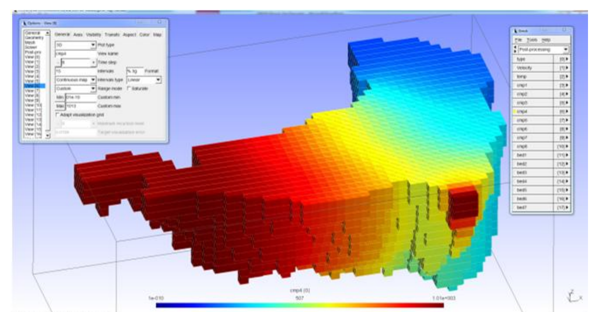
Clay



Cesium adsorbed by clay



Dissolved cesium



A reservoir is very effective to reduce cesium migration downstream

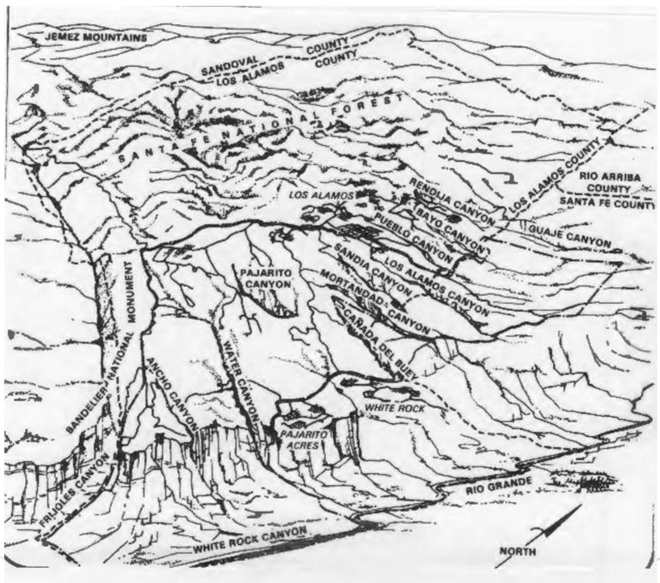
Simulation of the Ogi Dam Reservoir with 3-D FLESCOT

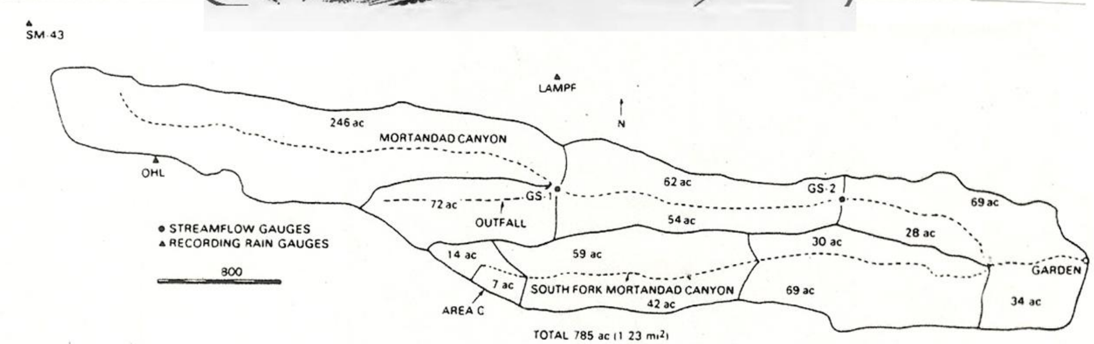
Purpose: Assess Potential Risk of Plutonium Migration
Combined Biological and Physical Radionuclide Transport

Joint Study with LANL

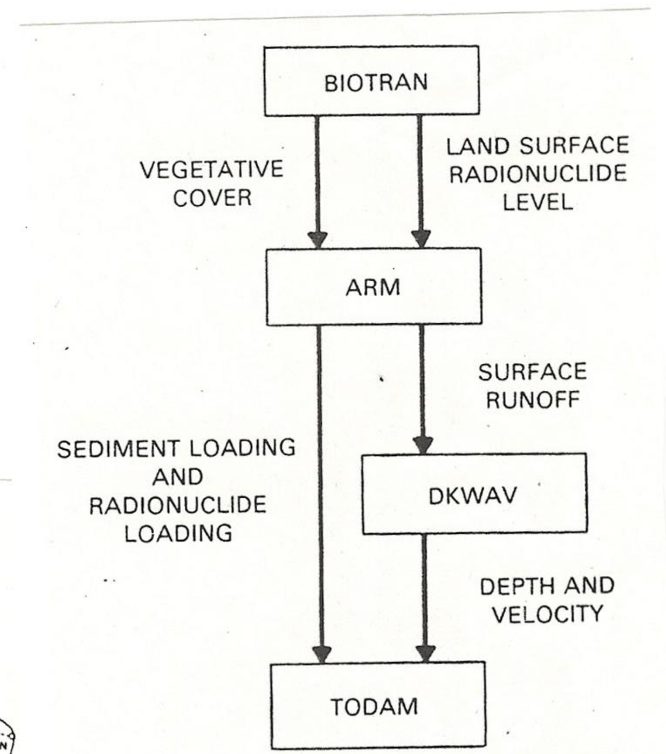
Plant growth/uptake/death → Deposit radionuclides on soil surface in a more easily erodible form → transported by wind and water

Los Alamos National Laboratory's
Mortandad and South Mortandad Canyons





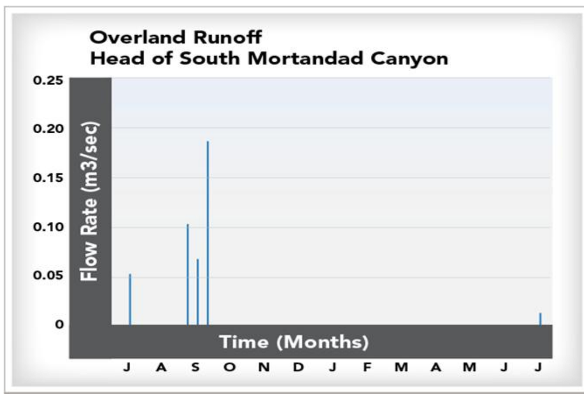
Interactions of Biological and Physical Mechanics



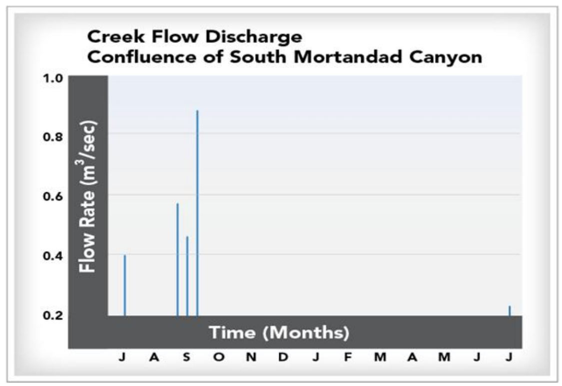
Plutonium Transport, Deposition and Re-suspension in South Mortandad Canyon with TODAM

- Almost all Pu is transported by sediment
- Clay contained the highest Pu concentration, then silt and sand
- Sand Consists of most of sediment in the stream
- Need to track movements of each sand, silt and clay to evaluate migration of Pu

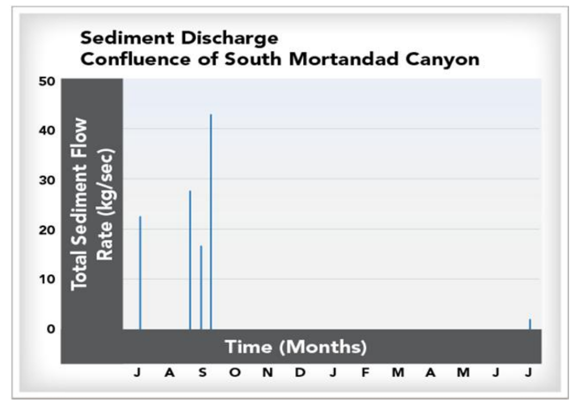
Overland runoff



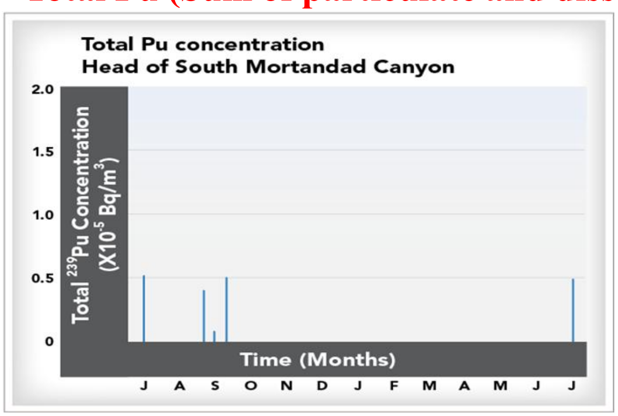
In-Stream water discharge



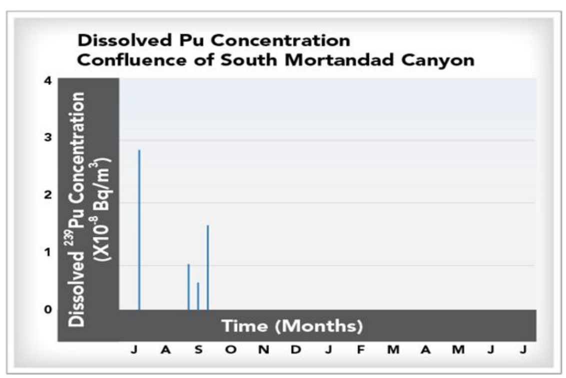
In-stream Sediment discharge



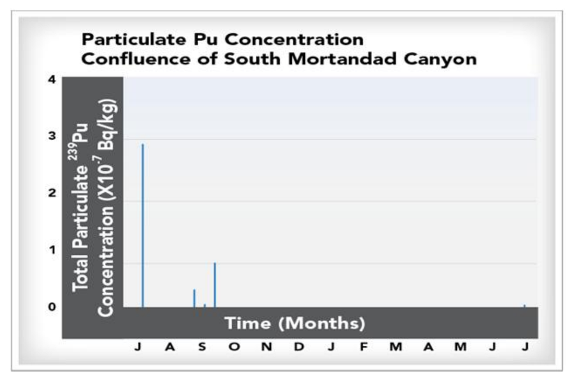
Total Pu (Sum of particulate and dissolved)



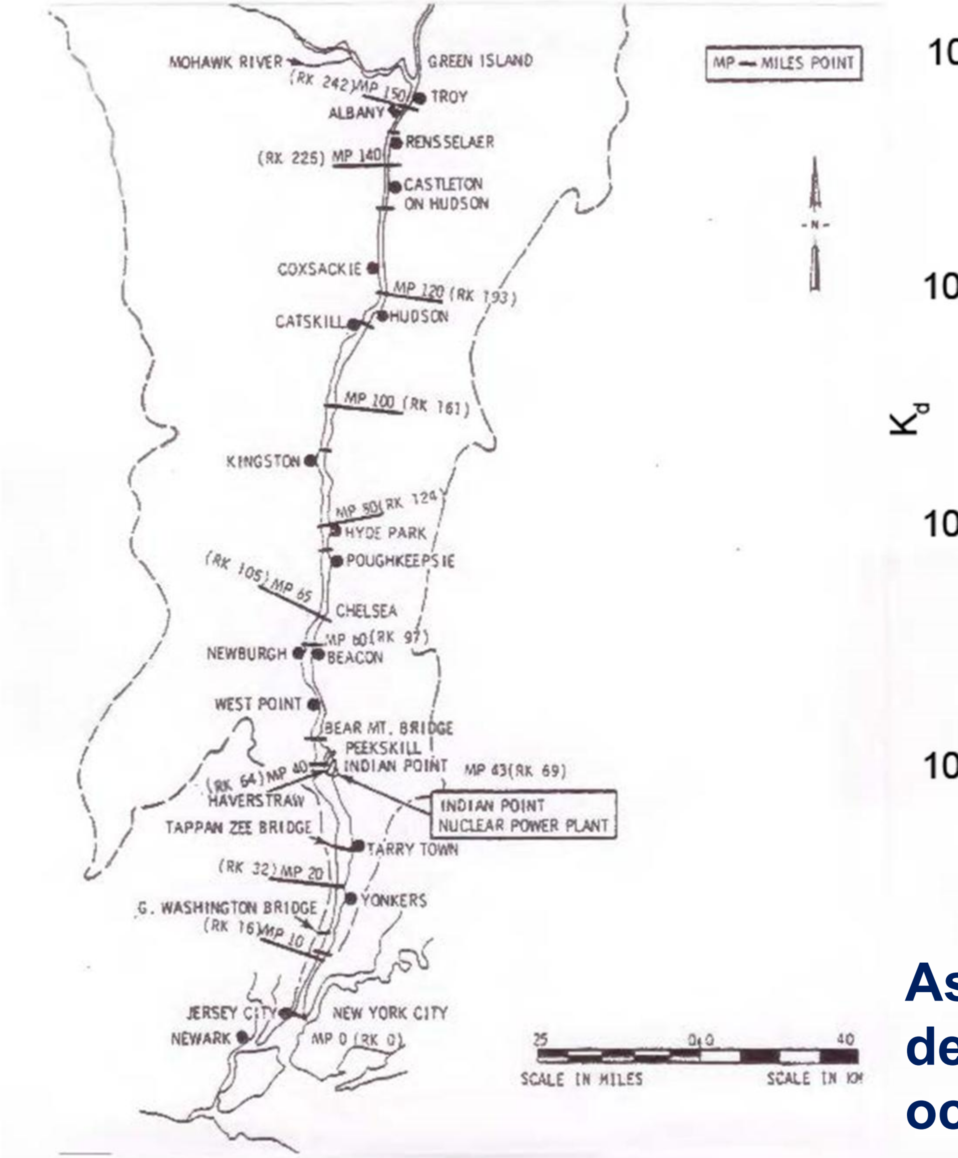
Dissolved Pu

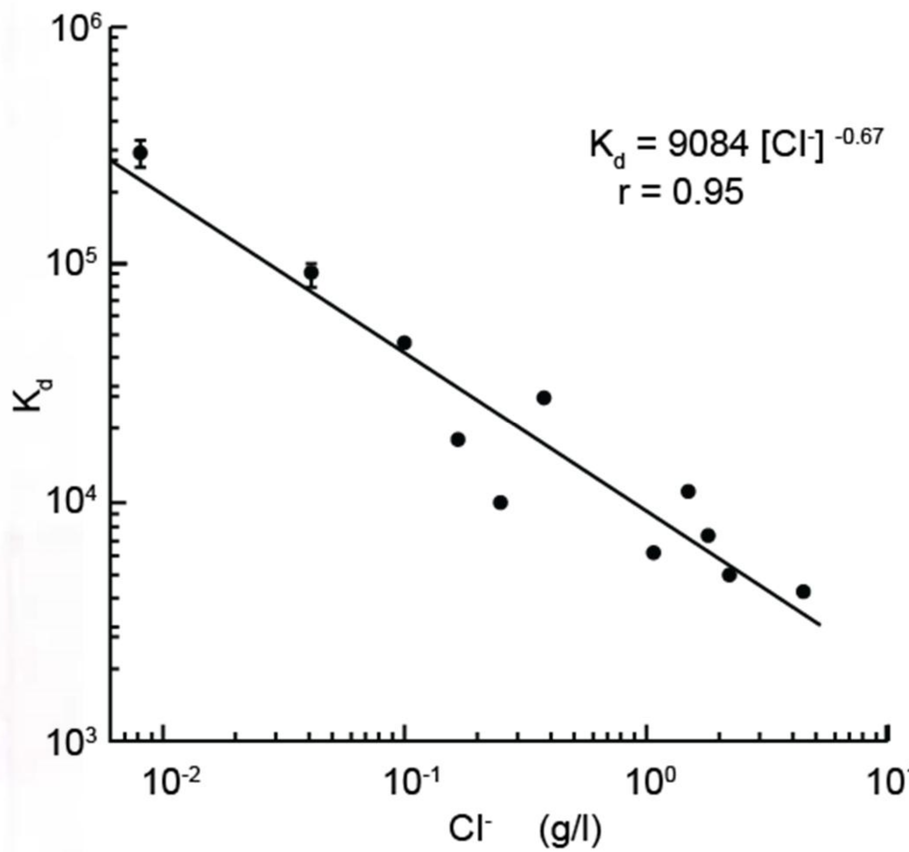


Particulate Pu



Purpose: Assess ^{137}Cs Spill to the Hudson River
Hudson River Modeling with 3-D FLESCOT Code
^{137}Cs Kd value affected by salinity was measured in the river

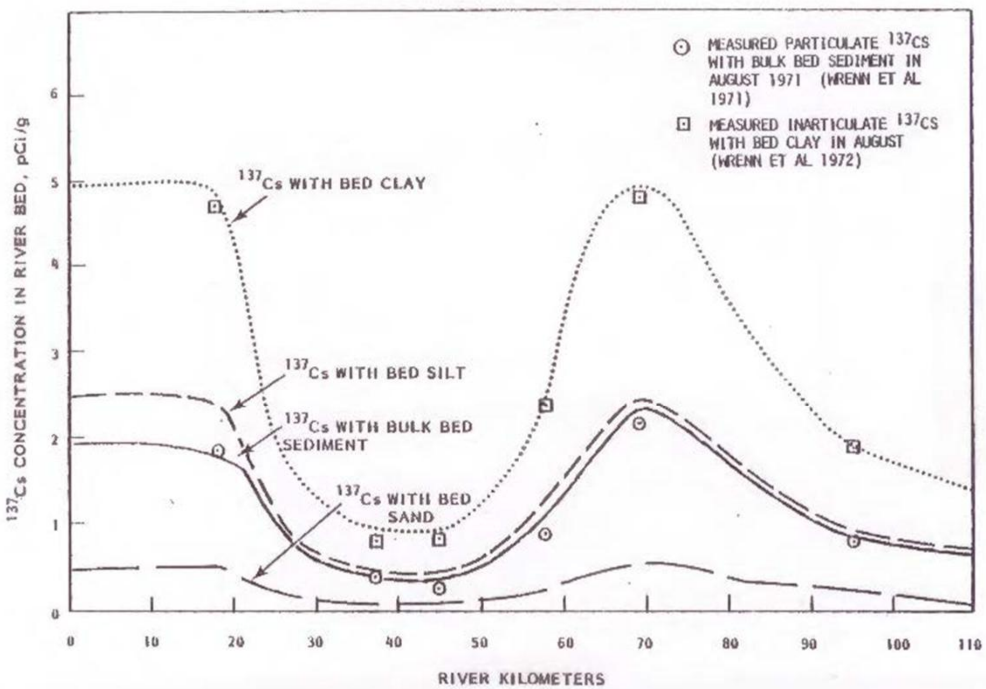




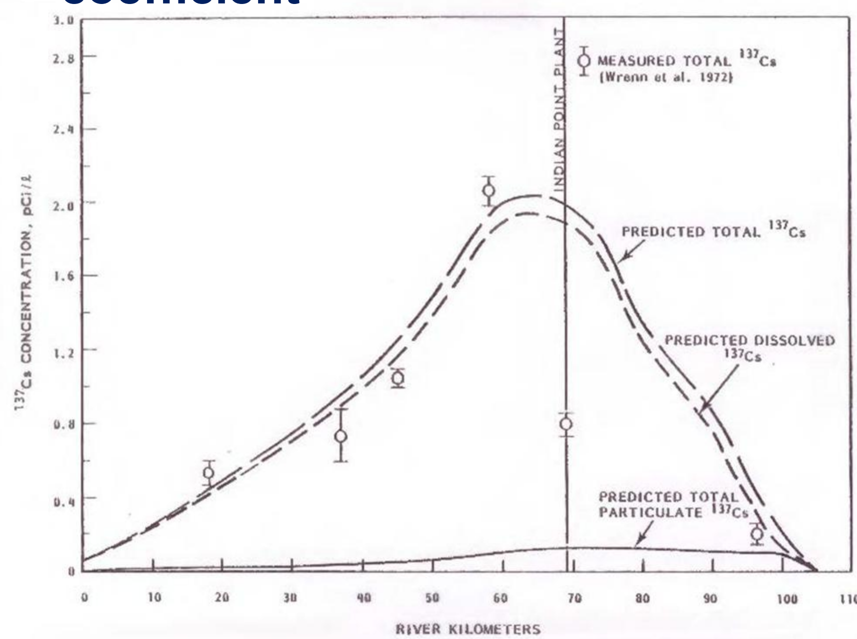
As ^{137}Cs approaches New York City, desorption of ^{137}Cs desorption occurs

FLESCOT Simulation of ^{137}Cs Migration

FLESCOT is **Validated** with the Data



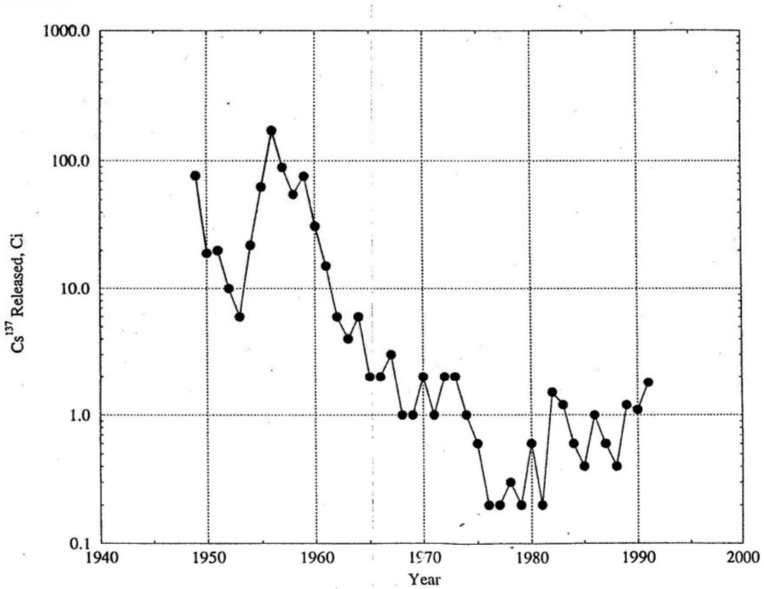
- **Importance of Clay on ^{137}Cs migration**
- **Main river sediment is sand, as most rivers are**
- **Need to simulate sand, silt and clay separately**
- **Importance of sediments depends on sediment concentrations and magnitude of distribution coefficient**



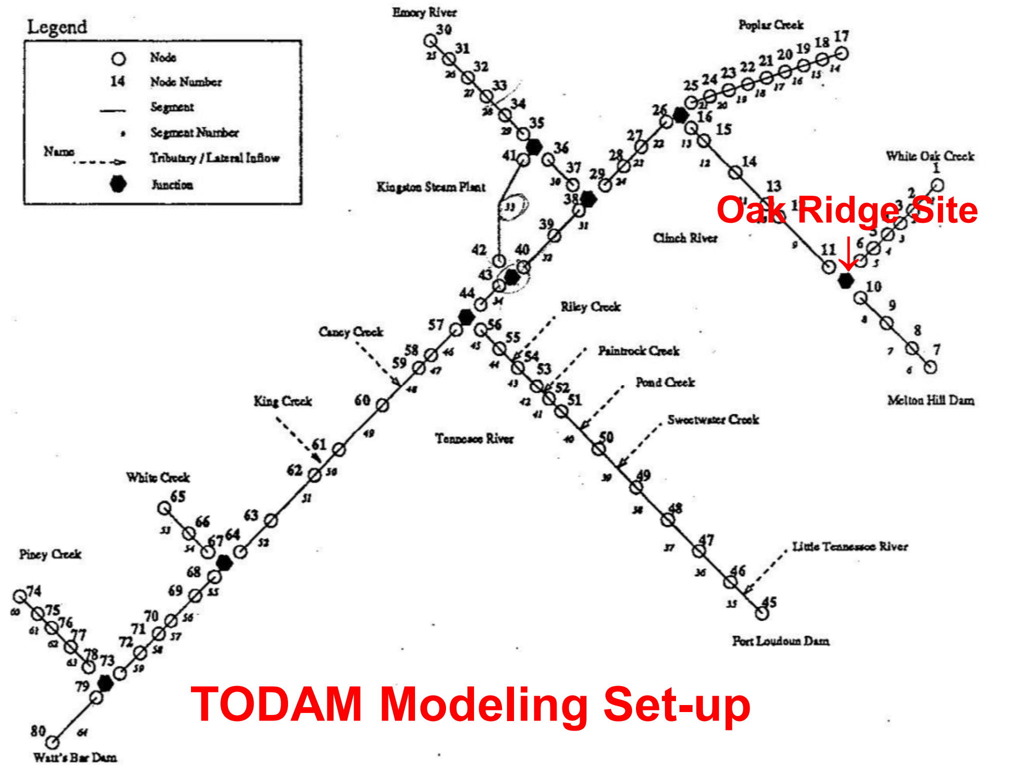
Approaching New York City,

- Suspended silt and clay deposit to the river bottom through flocculation
  - Forms long-term source of contamination
  - ^{137}Cs: Desorb from sediment
  - Relatively dissolved ^{137}Cs increases

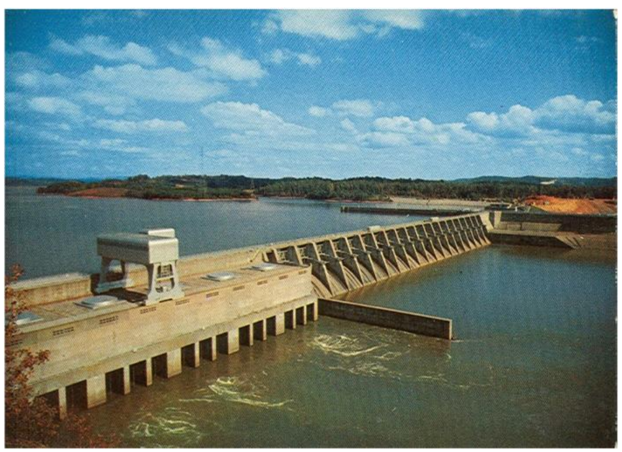
Purpose: Evaluate Potential Need for River Remediation
Clinch and Tennessee River Contamination by ^{137}Cs Releases from Oak Ridge Site over a Half Century

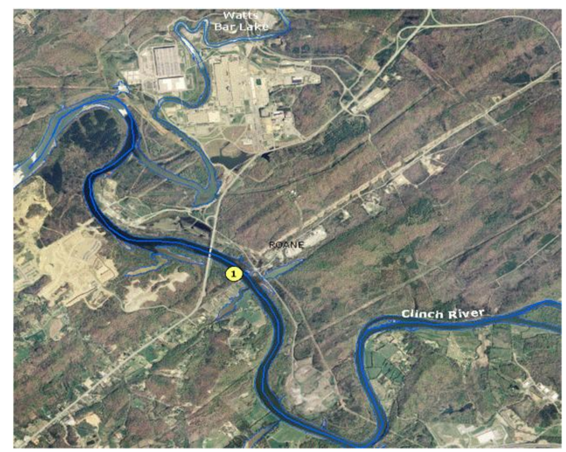


^{137}Cs Release from Oak Ridge Site



TODAM Modeling Set-up

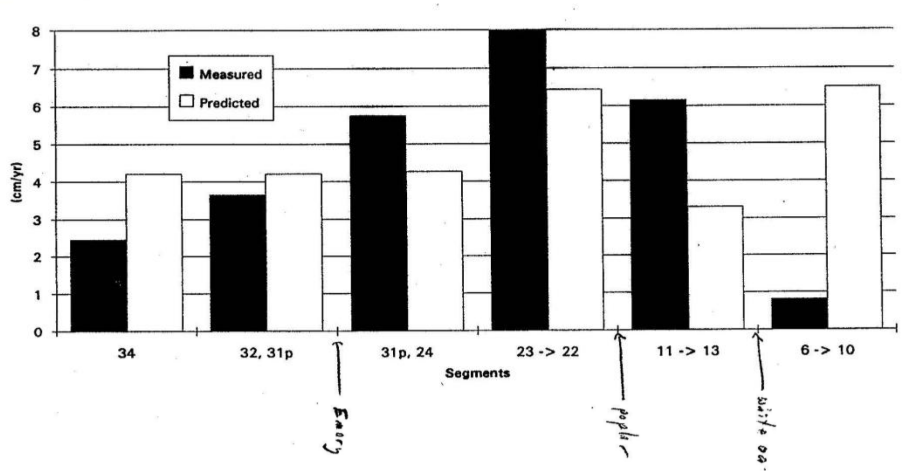


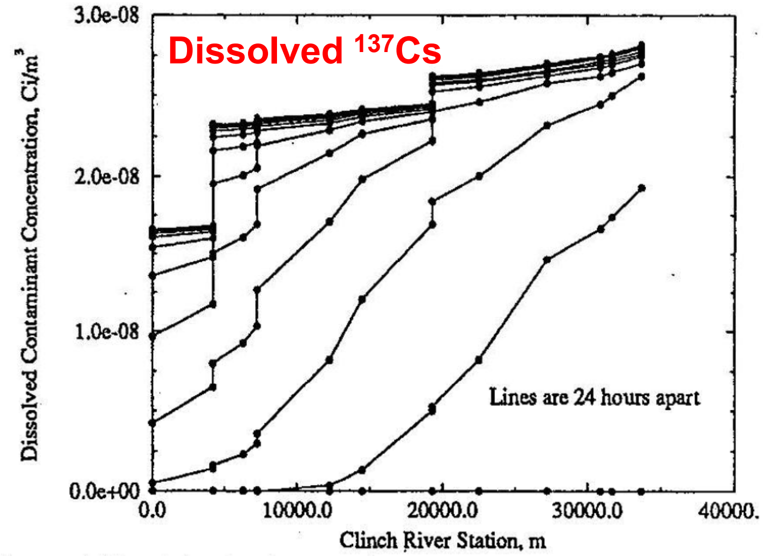


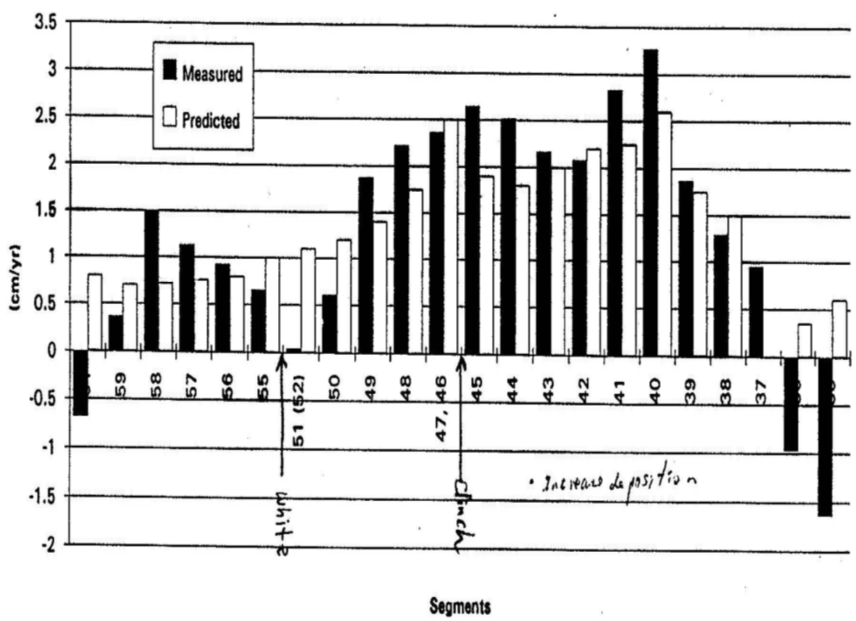
The Clinch and Tennessee River Sedimentation Rates over Decades: ^{137}Cs Prediction in these rivers

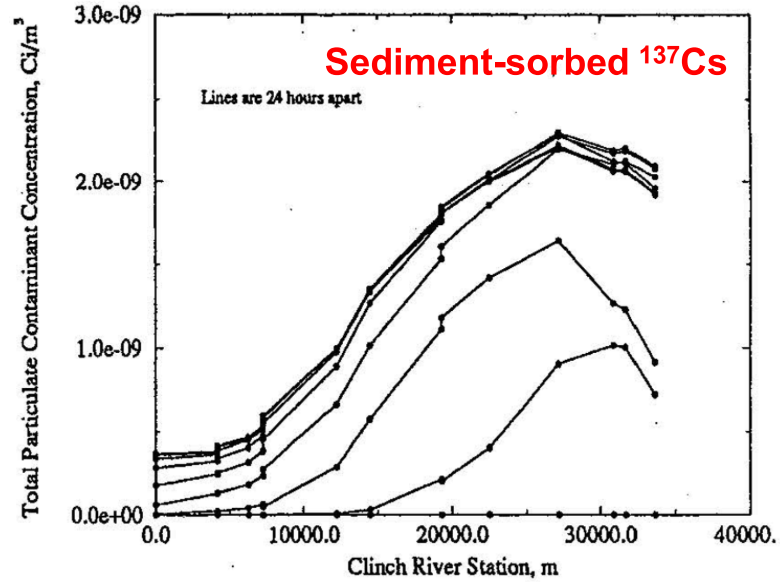
Joint Study with ORNL

TODAM was Validation









Main Aquatic Remediation Methods and Their Effectiveness

Intended Water Body	Radiation method	Radionuclides	Decontamination Factor	Disposal Need
Surface water	Zeolite	^{134}Cs, ^{137}Cs, ^{90}Sr	90% and greater	Yes
Surface water	Prussian Blue	^{134}Cs, ^{137}Cs	90% and greater	Yes
Surface water	Lime	^{90}Sr	Variable up to over 90%	Yes
Surface water	Settling Pond and Reservoir	Suspended radionuclides in rivers	Variable	No*
Surface water	Dredging	Deposited radionuclides in rivers and coastal water	Variable	Yes
Surface water	Diversion waterways	All radionuclides in rivers	100%	No*
Surface water Groundwater	Block radionuclide influx to water	All radionuclides in land surface, or within the surface water	100%	Yes/No*

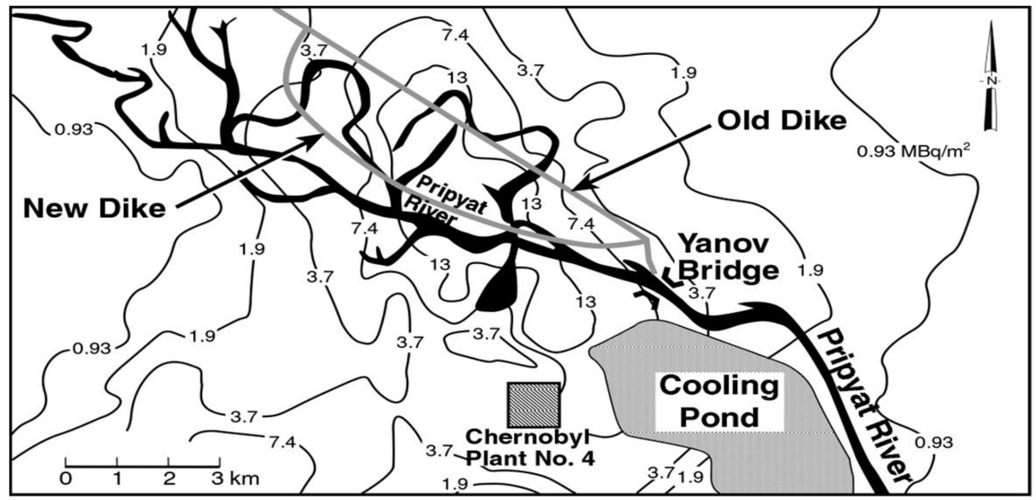
23

Do nothing – Natural attenuation

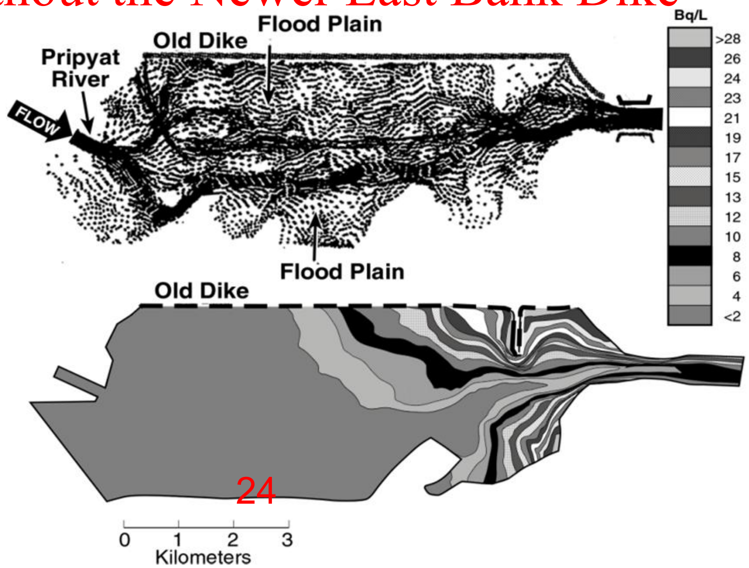
Purpose: Reduce 90Sr Concentration Level in the Dnieper River

- **50% of ^{90}Sr comes from Chernobyl Pripyat River Floodplain**
- **Remediation Effectiveness Assessment With FETRA Code**

Solution: Block off Contaminated Floodplain with a Dike

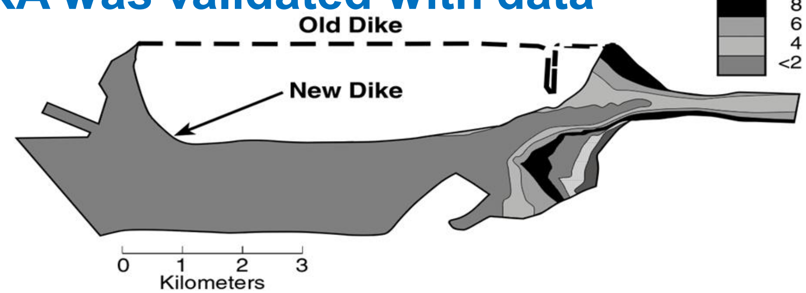


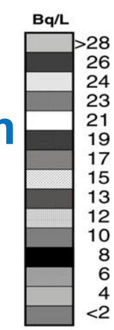
Without the Newer East Bank Dike



With the Newer East Bank Dike

- **The dike construction actually reduced aquatic pathway dose from ^{90}Sr by 50% caused by flooding**
- **FETRA was validated with data**



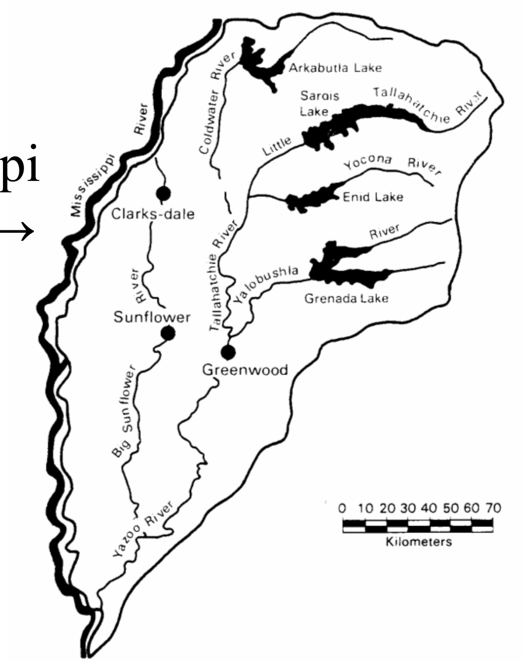


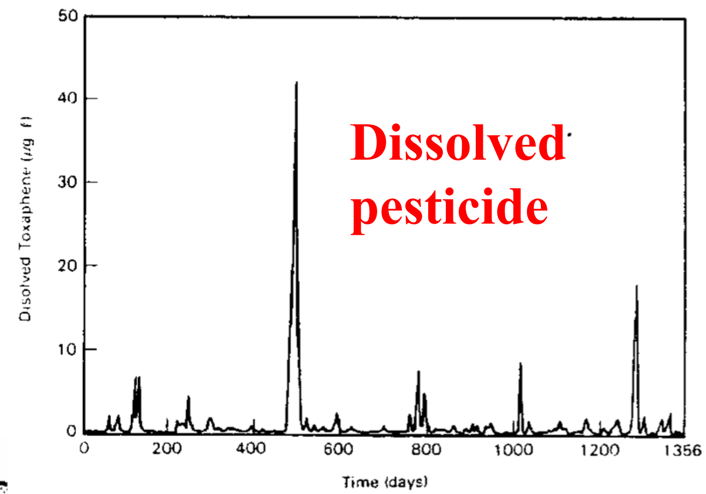
Purpose: Determine if the pesticide should be banned in U.S. Use of Models to Make this Decision

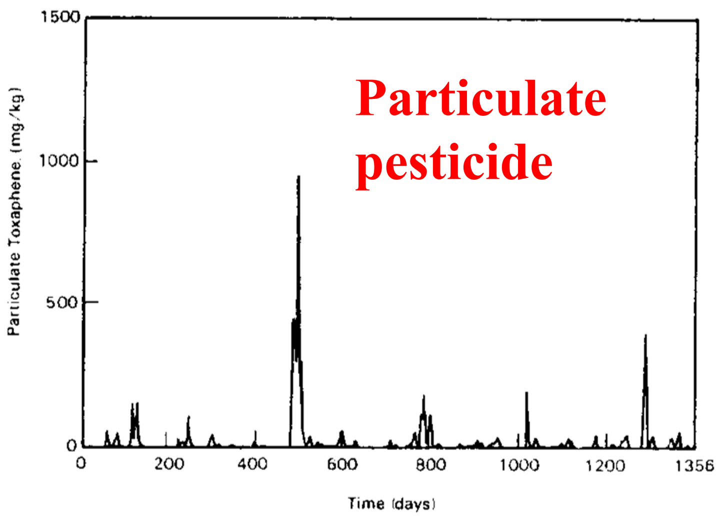
Solution: EPA decided to ban the pesticide with this modeling result

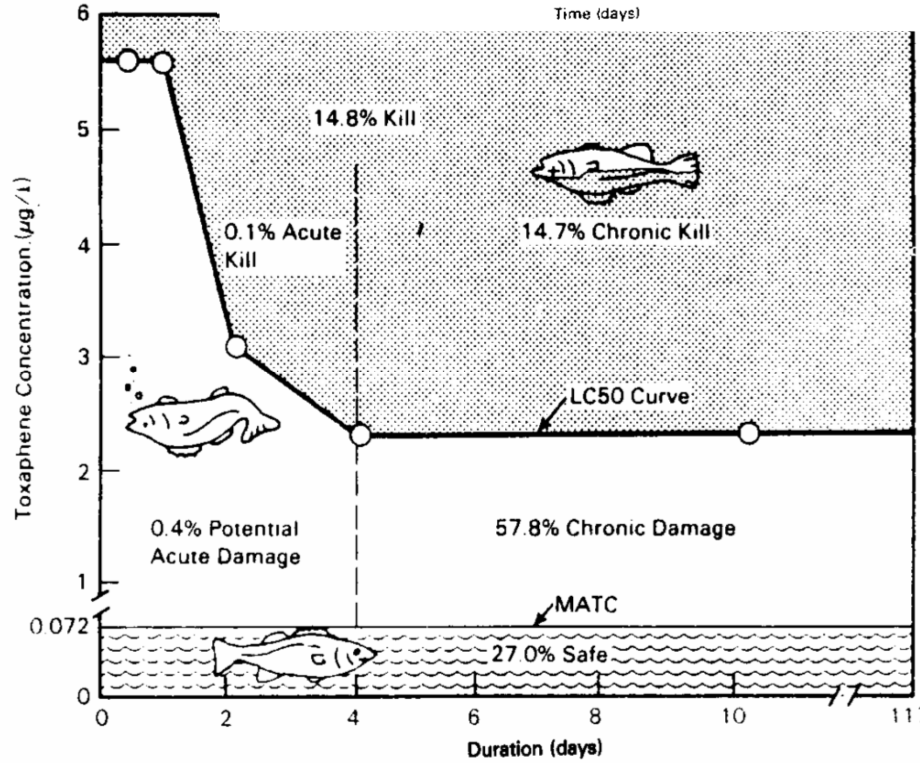
Watershed of Yazoo River and its Tributaries

Mississippi River →





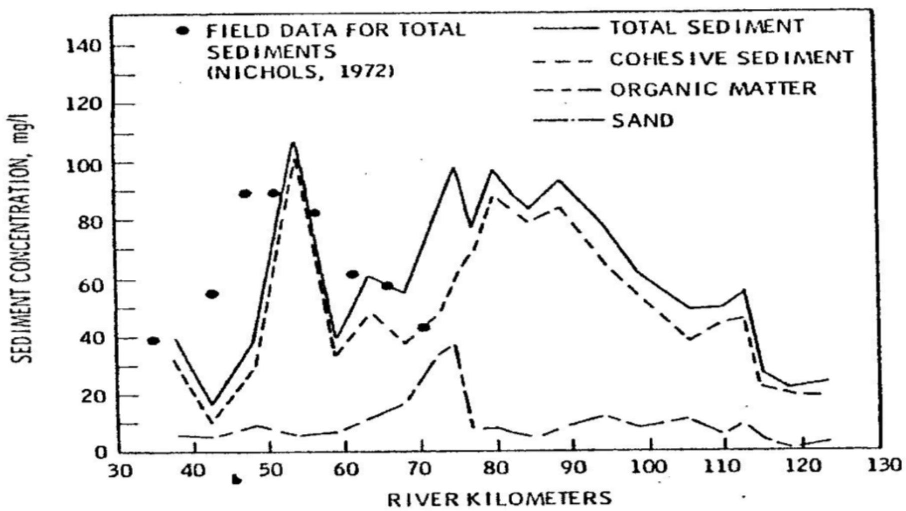




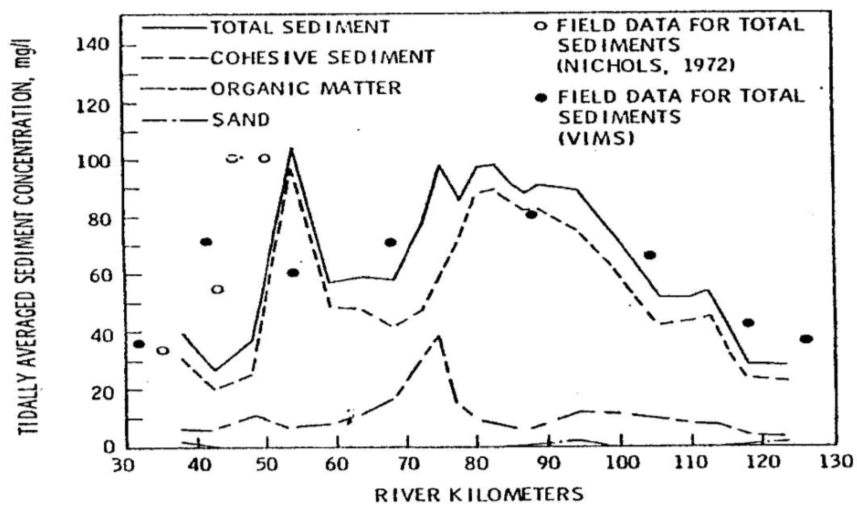
Commercial Fishing was banned
Purpose: Where to dredge and its effectiveness to restore fish?
Simulation of Sediment and Pesticide Transport with 2-D FETRA Code

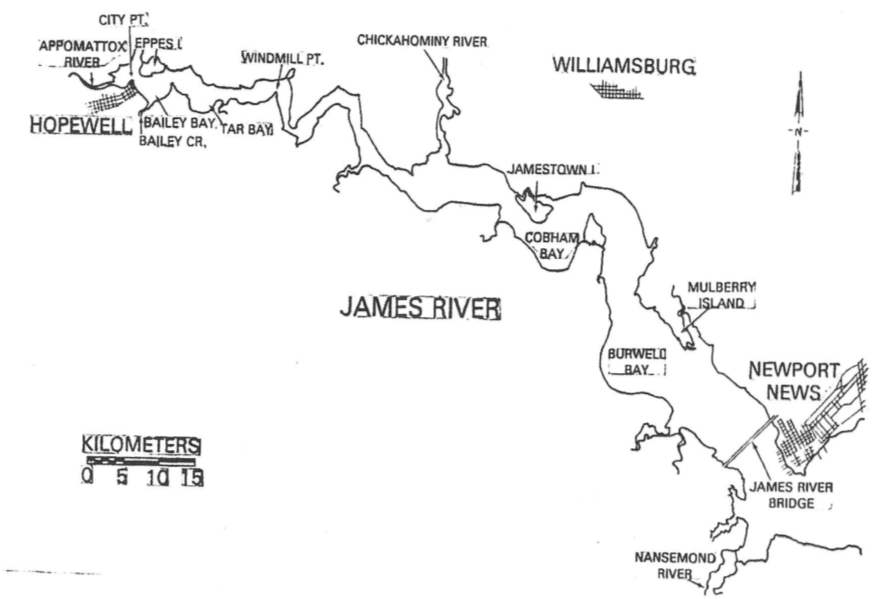
Sand, Cohesive Sediment, Orgasmic Matter under Tidal Flow with Net Freshwater River Discharge of 243m^3/s

Tidally Varying Sediment Concentration at Slack Tide

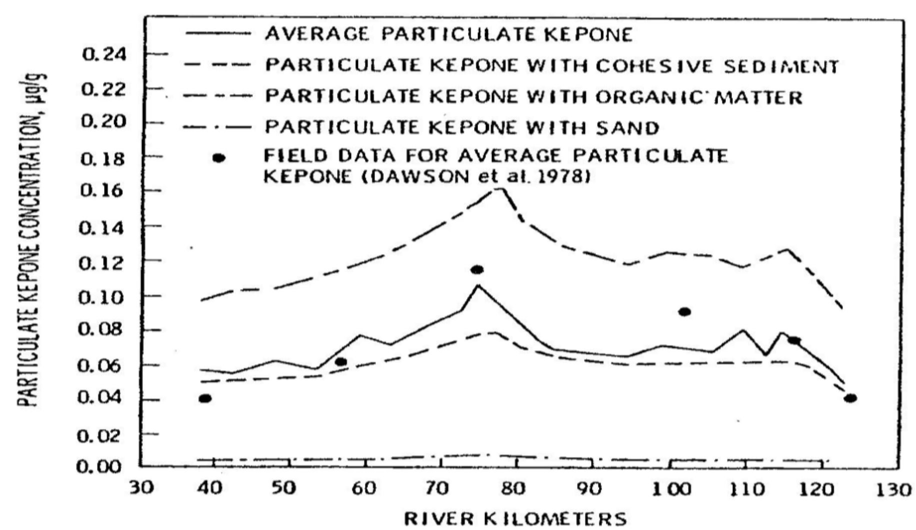


Tidally Averaged Sediment Concentration

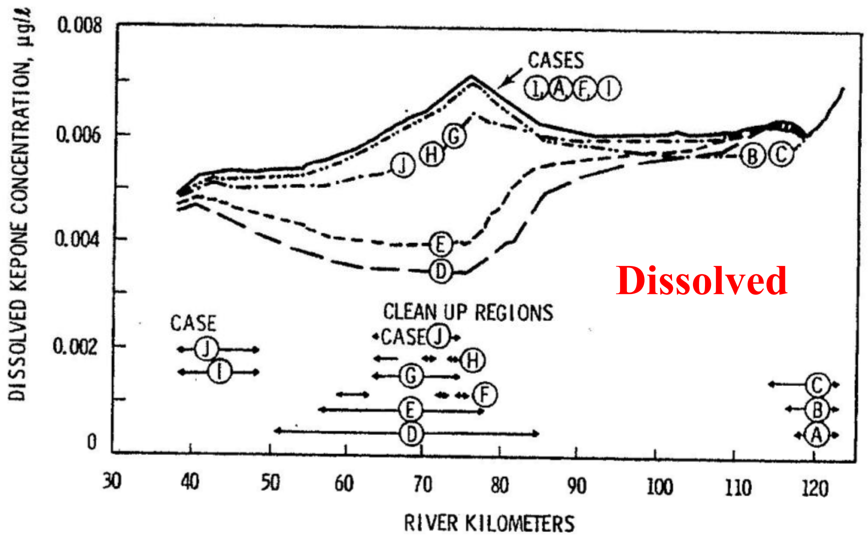


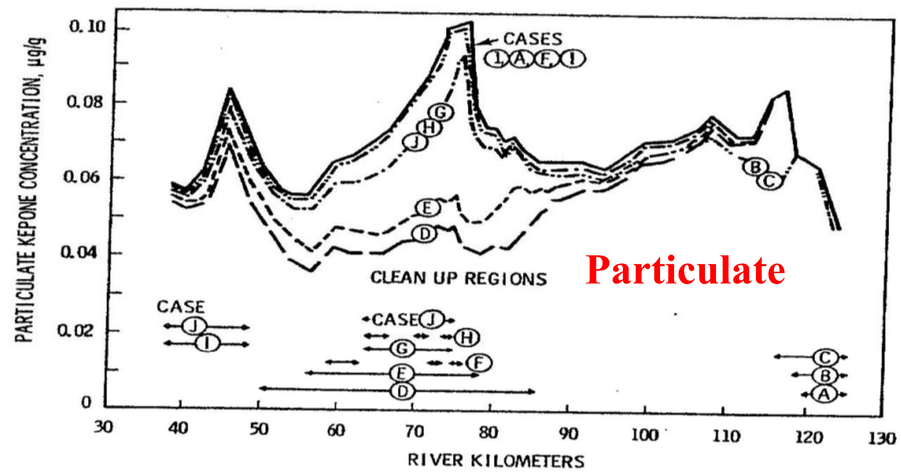


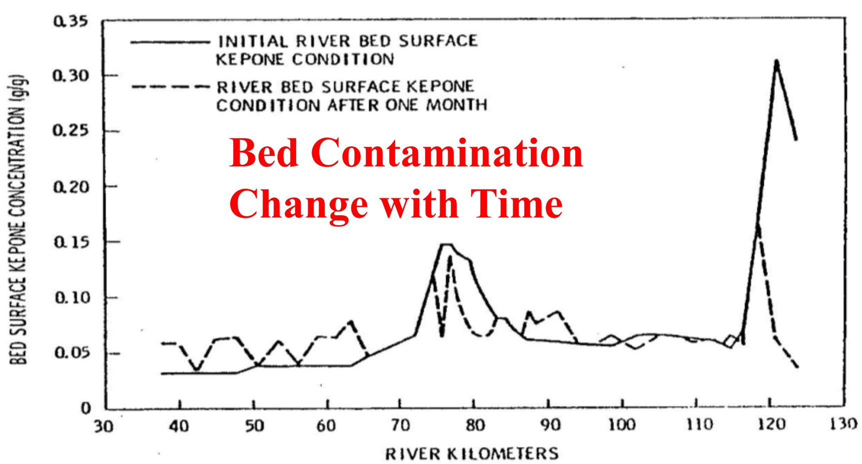
Particulate Pesticide Concentration at Maximum Ebb Tide

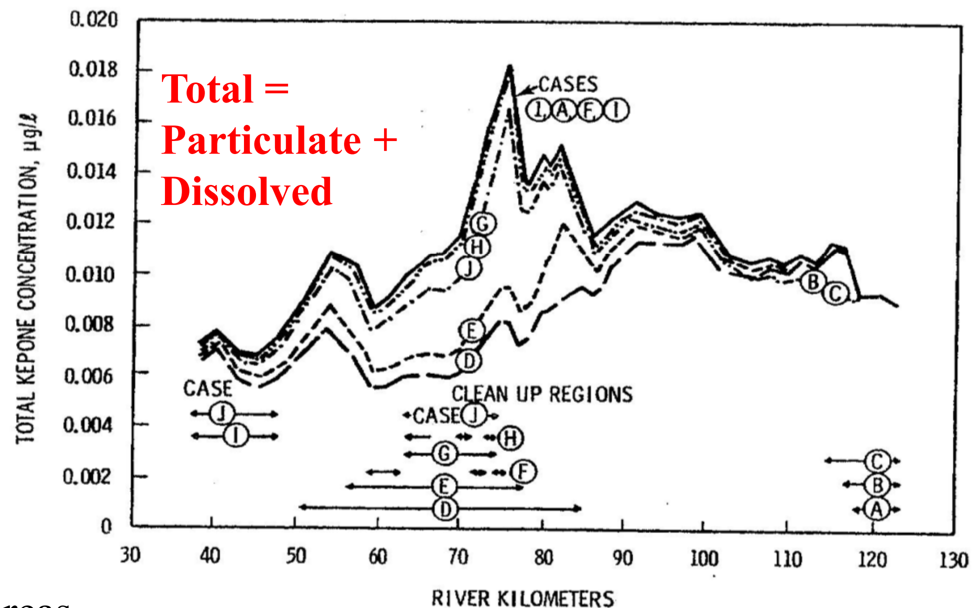


Remediation Effectiveness due to Dredging (2-D FETRA Code）









Modeling Conclusions

- Requires dredging of large areas
- Natural cleaning is effective to reduce contamination level

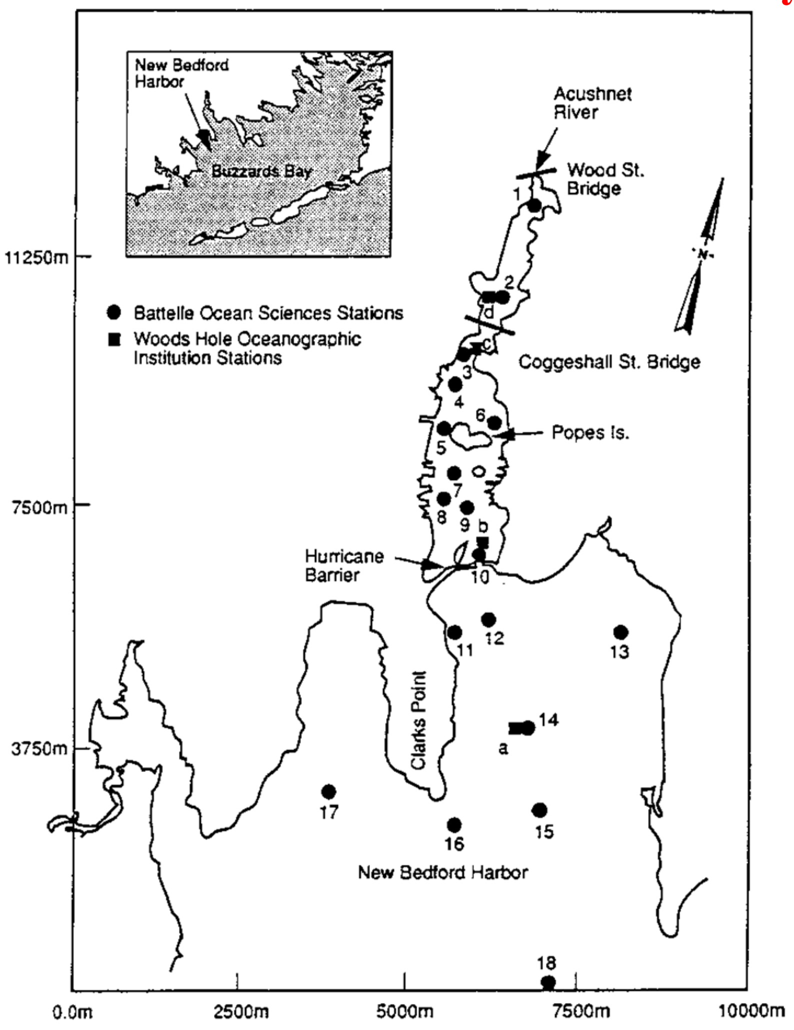
Solution

- No river remediation
- Commercial Fishing is back, as predicted by the model

Purpose: Decide Where to Dredge and clean-up effectiveness to restore fish and lobster
Remediation Assessment with 3-D FLESCOT Code
Dredging Bottom Sediment Contaminated by PCBs and Heavy Metal

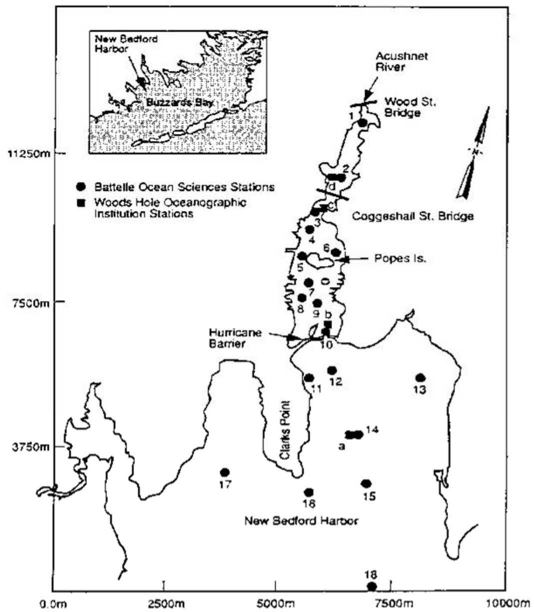
Aquatic biota mainly take up contaminant through foods → Assessment of food web

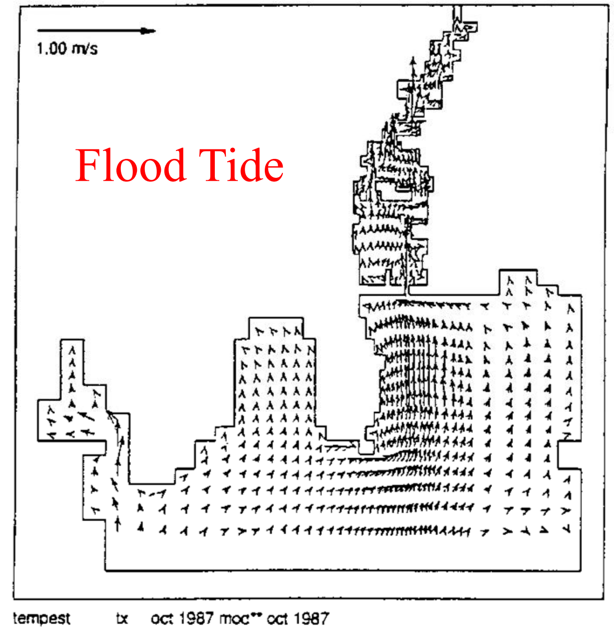
New Bedford Harbor Buzzards Bay

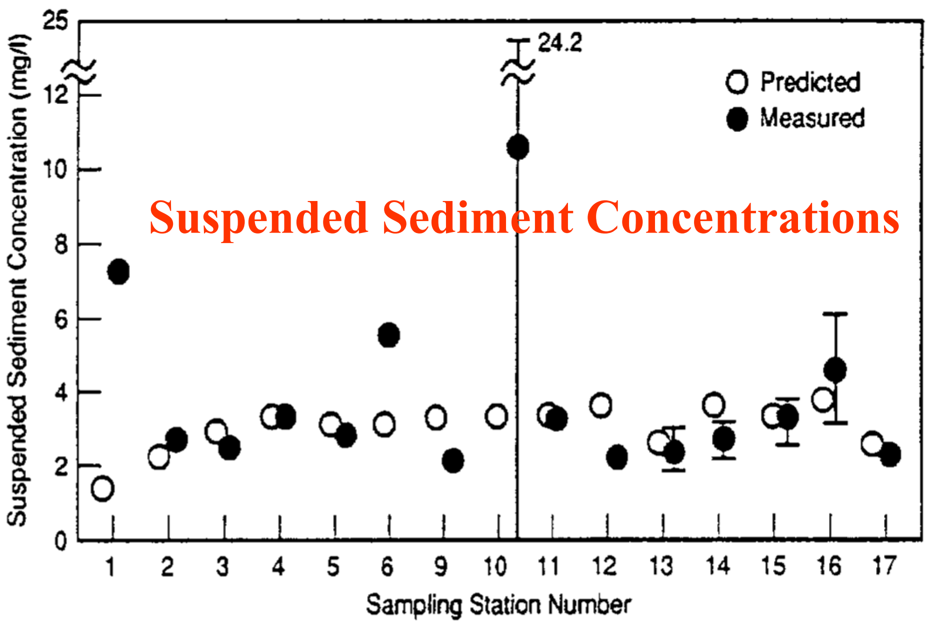


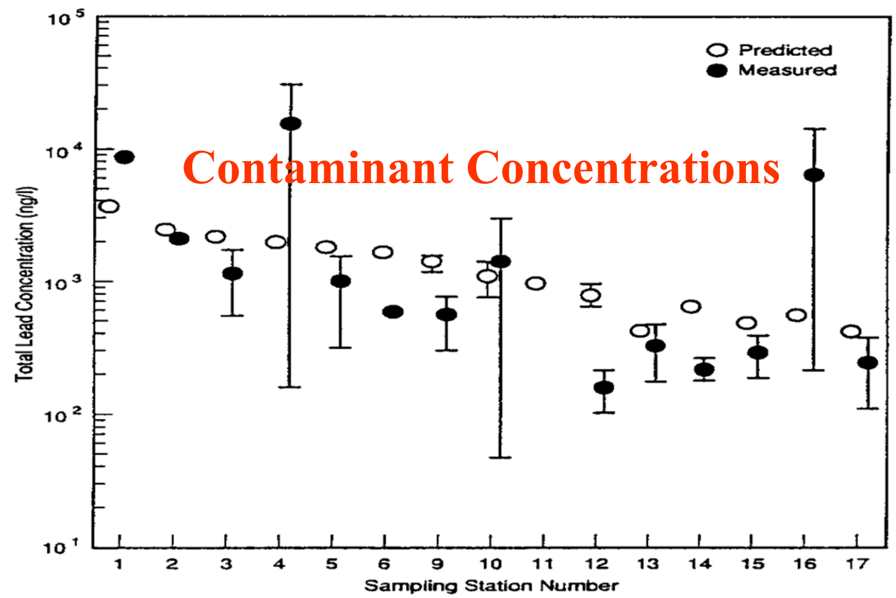
FLESCOT Application to New Bedford Harbor PCB and Heavy Metal Contamination

New Bedford Harbor and Buzzards Bay



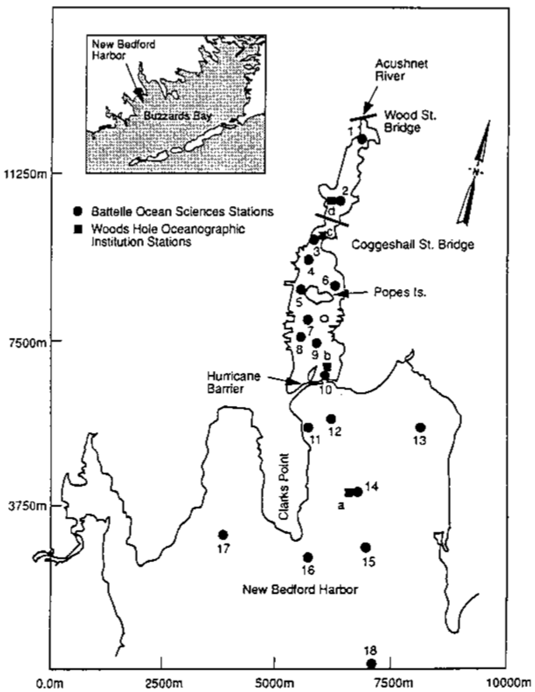




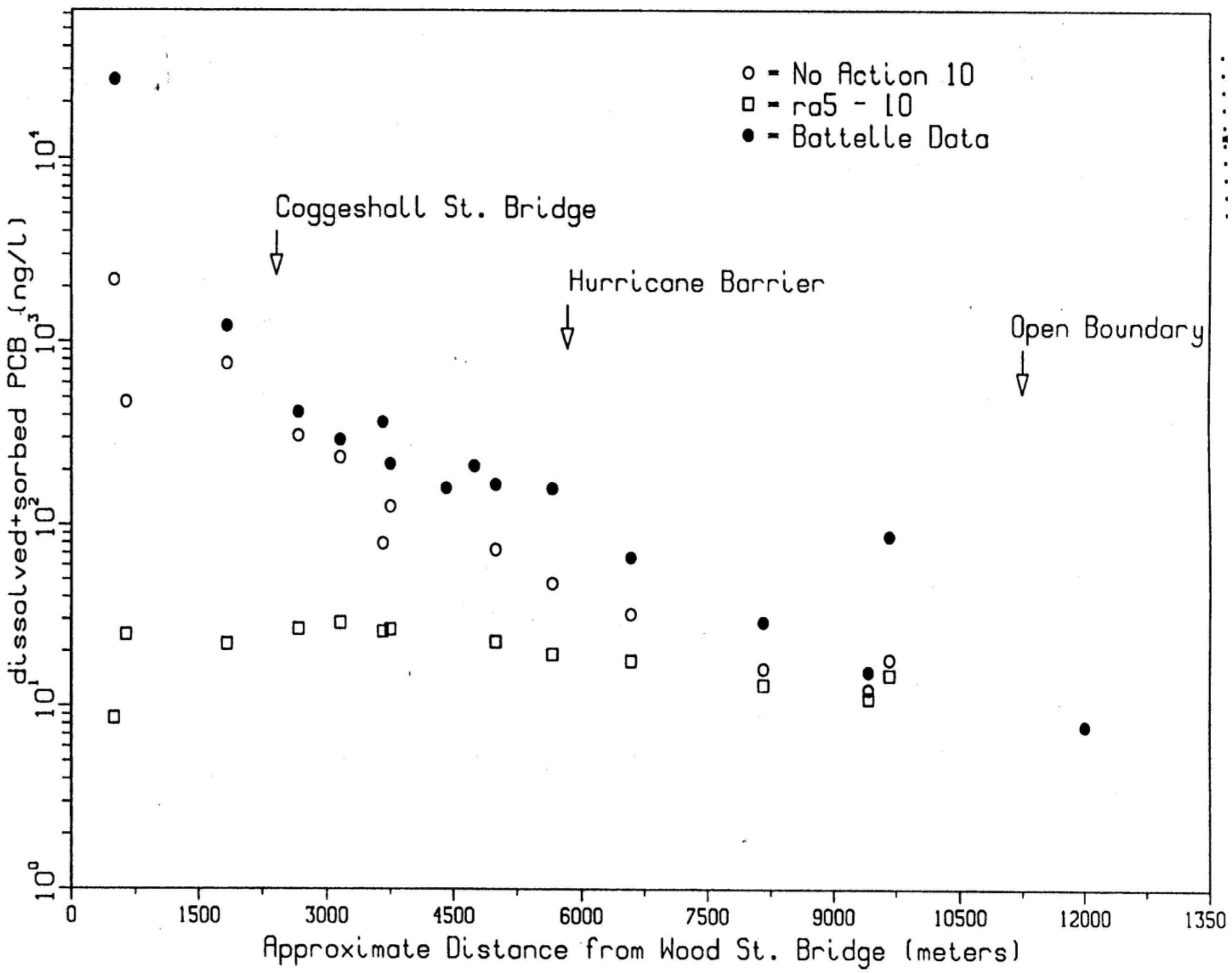


Remediation: Dredging Bottom Sediment in Lower Estuary

Prediction: PCB Concentrations after 10 years with and without dredging



PCB Concentrations after 10 years

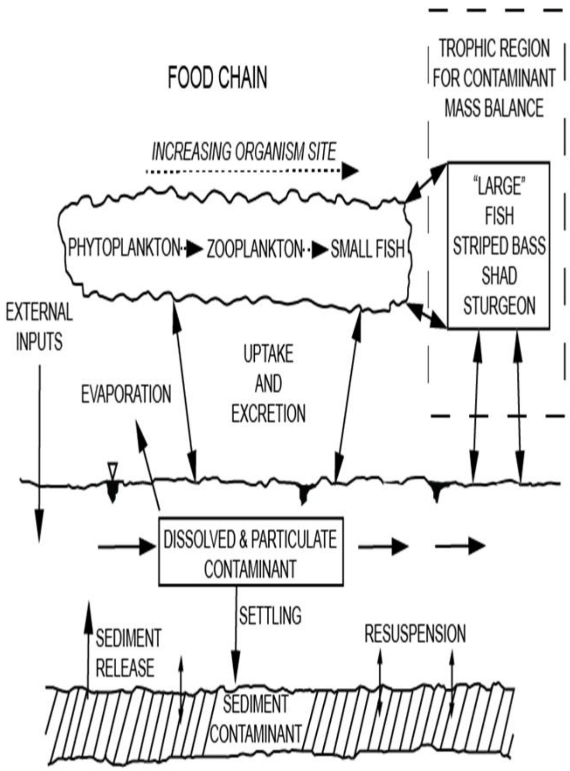


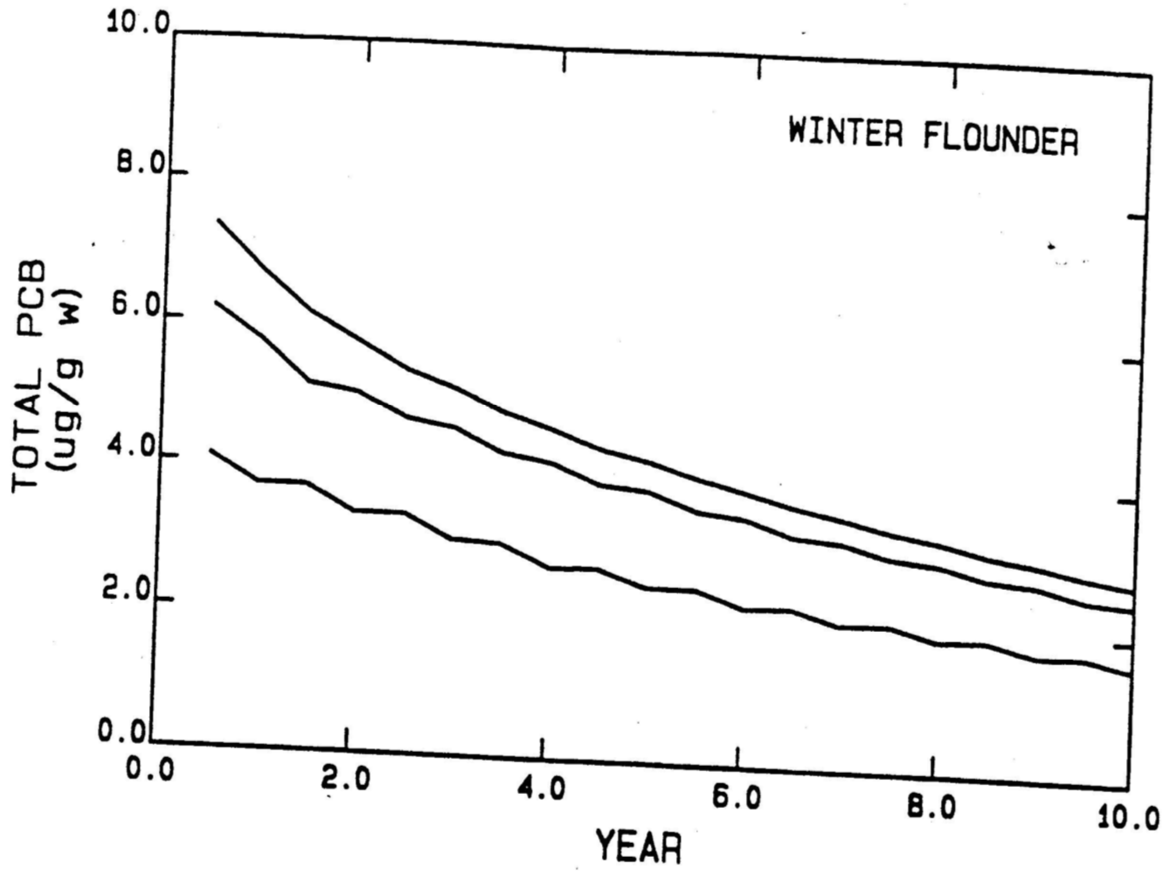
PCB Reduction in Winter Flounder in Lower Estuary with Lower Estuary Dredging

0-, 2-, 5-year old Winter Flounder's PCB Concentrations in Lower Estuary

- **65% more reduction without dredging**
- **USA FDA Action Limit: 11 µg/g**

Food Web Example





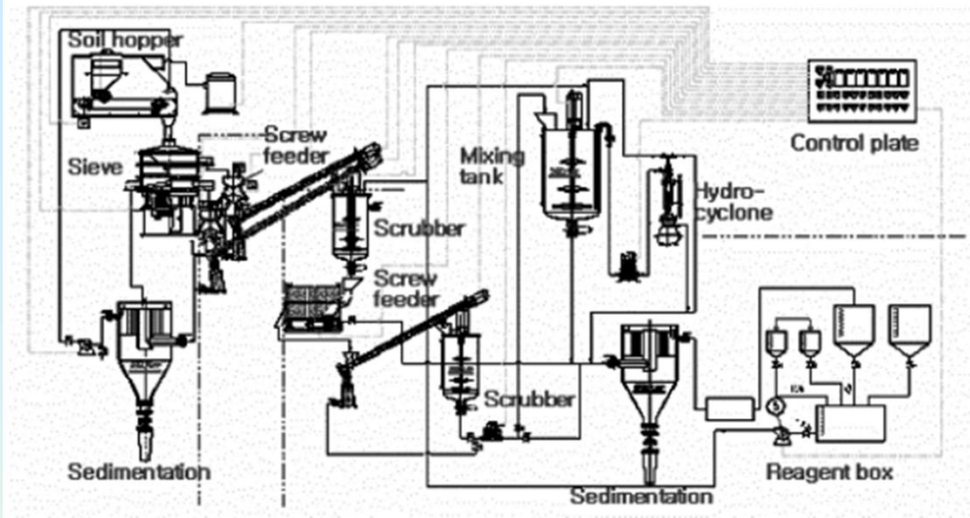
Some Remediation of Cesium-Contaminated Soil

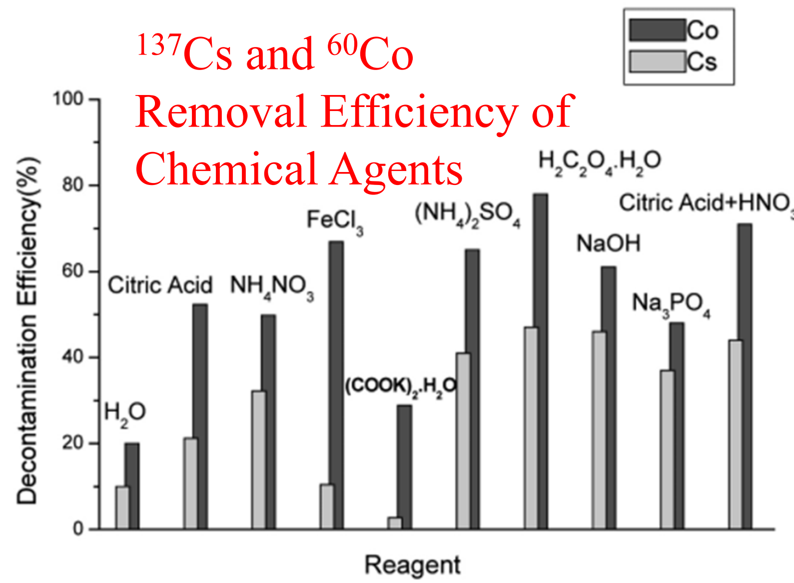
Technology	Description	Comment
Excavation	Scrape upper soil layer and either wash soil or dispose.	Effective, but remove valuable topsoil unless replaced with new or washed soil.
Isolation	Engineered cover	Isolates contaminated materials and reduces exposure
Grouting	Inject grout material to entrap the radionuclides in a monolith	Isolates radionuclides, but restrict future land uses
In-situ leaching	Leach with acid or ion exchange and a complexing agent, such as citrate	Applicable to shallow soil. Excess leachate must be collected. Risk of uncontrolled mobilization. Effectiveness depends on soil characteristics
Physical and radiological soil separation	Separate soils with high concentrations from soils with low concentrations	Mature technologies. Effectiveness depends on soil characteristics
Ex-situ soil washing	Extract cesium from solids by washing with water or suitable extraction solutions	Effectives depends on soil characteristics

Removal and Disposal of Hanford Contaminated Soil

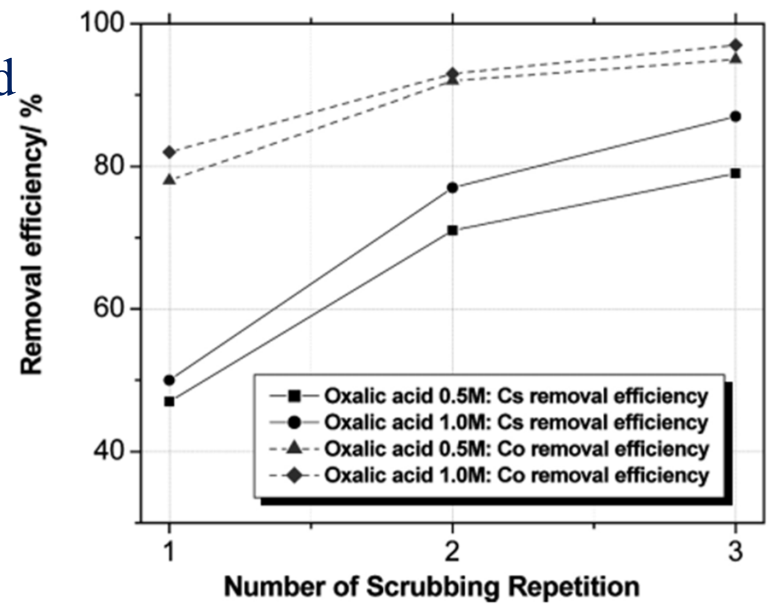
Soil Washing: Cesium Removal from Soil

Soil contaminated by TRIGA Reactor in Korea **G-N Kim et.al (2007)**





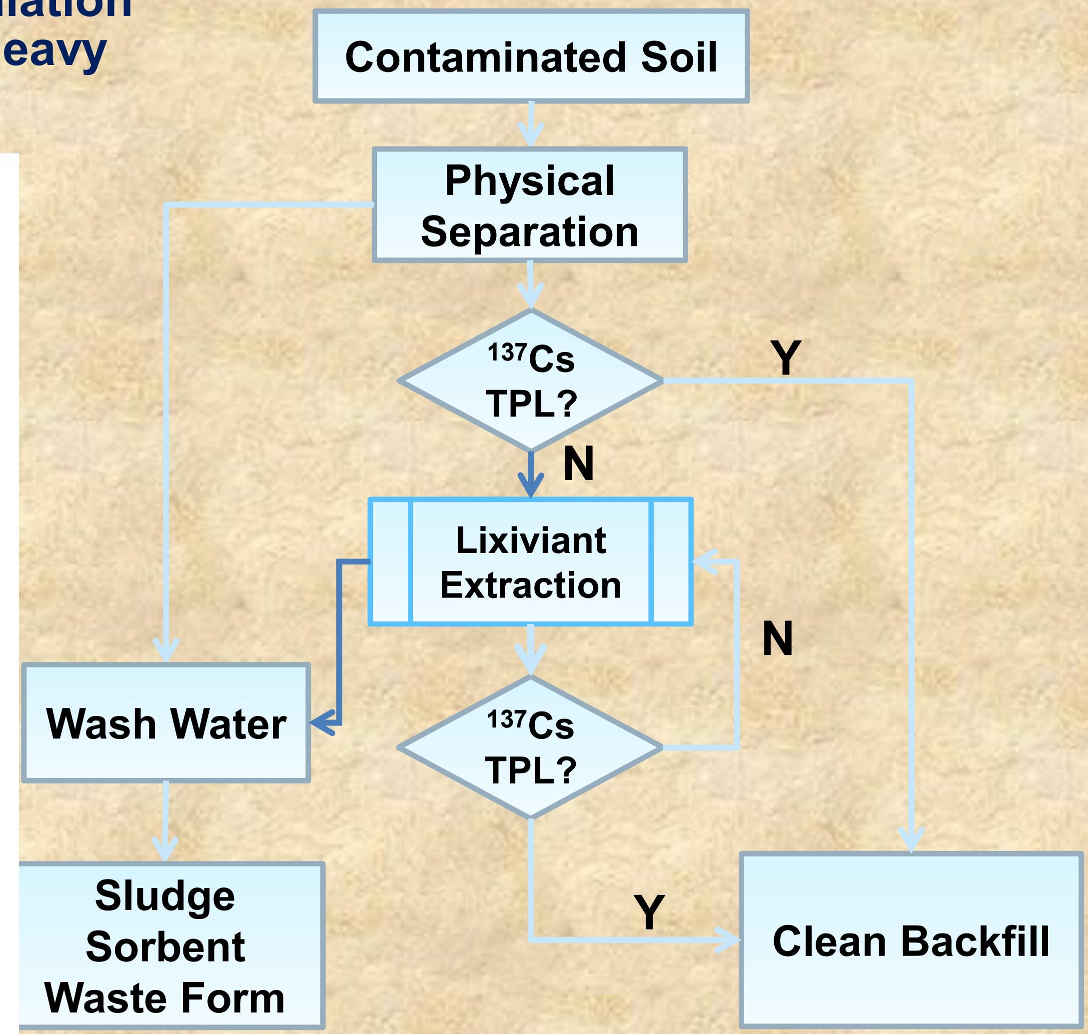
- Oxalic acid was the best to remove ^{137}Cs and ^{60}Co from the test sand up to 85% of ^{137}Cs
- Water removed 10% of ^{137}Cs from the sand
- Oxalic acid barely removed ^{137}Cs from silt and clay
- Fukushima silt and clay tightly adsorb ^{137}Cs and water does not desorb ^{137}Cs from Fukushima soil



^{137}Cs Contaminated Soils & Sediments
A Potential Ex-Situ Remediation

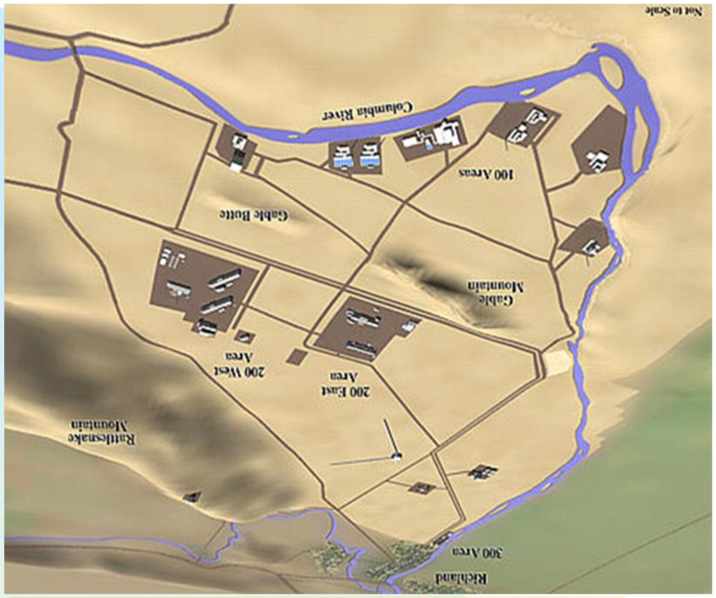
Soil Washing **– A Proven remediation technology for radionuclides, heavy metals & hydrocarbons**

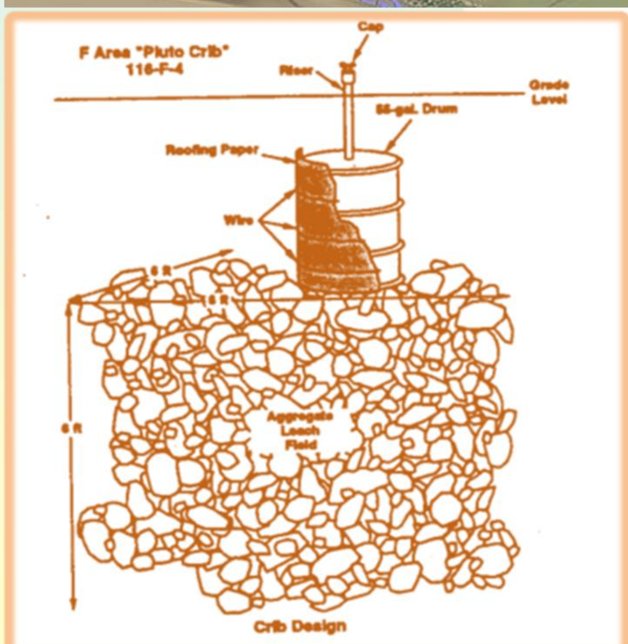
Soil Screening; Soil Feed Hoppers; Wet Screening; Arsenic Leaching; Leachate Clarification and As precipitation; Clean Sand Piles



TPL: Target Performance Level
(PNNL Lixiviant: nontoxic, and biodegradable)

^{137}Cs-contaminated Hanford Site Crib Soils Hanford Dominant Clay Mineral: Mica-type similar to Fukushima Soil to tightly Sorbs ^{137}Cs





Lixiviant Extraction Results

(Not-optimized)

2.00 – 0.25 mm Fraction Hanford Soil

Lixiviant Formal Conc	Initial ^{137}Cs Activity (Bq/kg)	Final ^{137}Cs Activity (Bq/kg)	^{137}Cs activity Reduction (%)
0.25	9.6E+03	2.7E+03	72
0.50	9.6E+03	2.1E+03	78
1.00	9.6E+03	1.6E+03	83
0.25	4.2E+03	1.5E+03	64
0.50	4.2E+03	1.0E+03	76

Extractions conducted at 96 ˚C for 6 hr

PNNL Lixiviant: nontoxic, and biodegradable

It has a potential to be applicable to Fukushima

Summary

Modeling:

- 1-d, 2-d, 3-d modeling have been used for
  - Aquatic environmental assessments of nuclear accidents, and past nuclear and industrial facility operations
  - Decision making of aquatic environmental remediation
- Modelers need to understand the physical and chemical phenomena they are trying to simulate
  - Cohesive sediment transport is important to determine radionuclide migration
- Modelers need to know what and how models predicts
- It is critical to conduct model calibration and validation
- Conduct modeling to predicts future contamination levels for many conditions including
  - Normal conditions
  - Not yet-occurred extreme weather conditions (e.g., severe storms and drought)
  - Remediation schemes before, during and after remediation

^{137}Cs removal from Soil:

- Soil washing with carefully selected chemicals is feasible to remove ^{137}Cs from soil, as Hanford Site has done
- Otherwise, excavation and capping of contaminated soil are commonly implemented in U.S.