Environmental Impact of Nuclear Waste
Insights from US Nuclear Weapon sites
What Can We Learn from Existing Nuclear Contamination?

Haruko Wainwright

03/09

BERKELEY LAB

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Nuclear Waste and Contamination

John Oliver, August 20, 2017

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Nuclear Waste and Contamination

"Nuclear Waste" Sites: Former Nuclear Weapon Production Sites
Deadly Plutonium: Half life of 24000 years

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Nuclear Waste and Contamination

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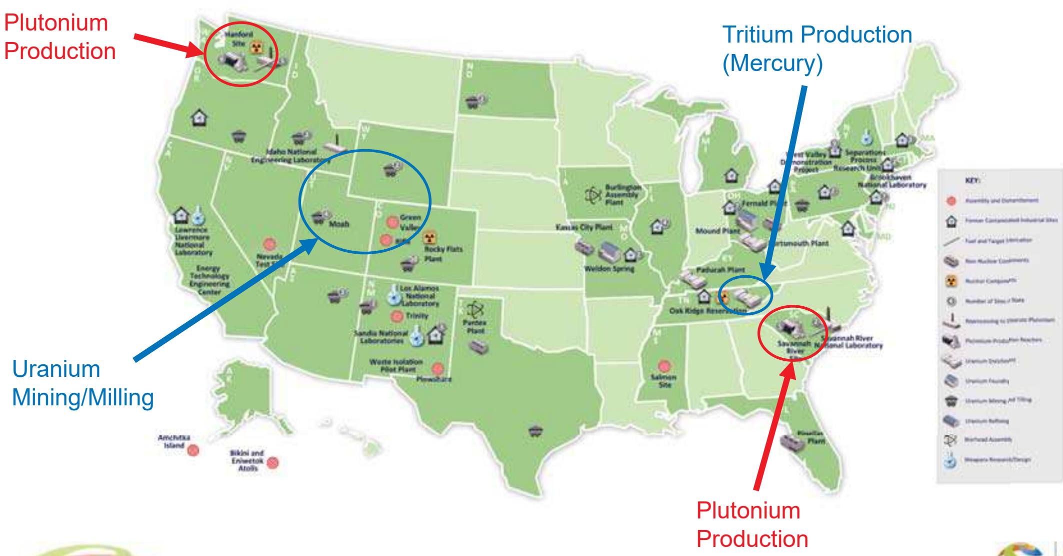
Nuclear Weapon Production Sites

Nuclear Weapon Production Sites



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Waste Disposal: Early Days

Savannah River Site F-Area

Rifle, CO

Hanford 300 Area

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Waste Isolation?

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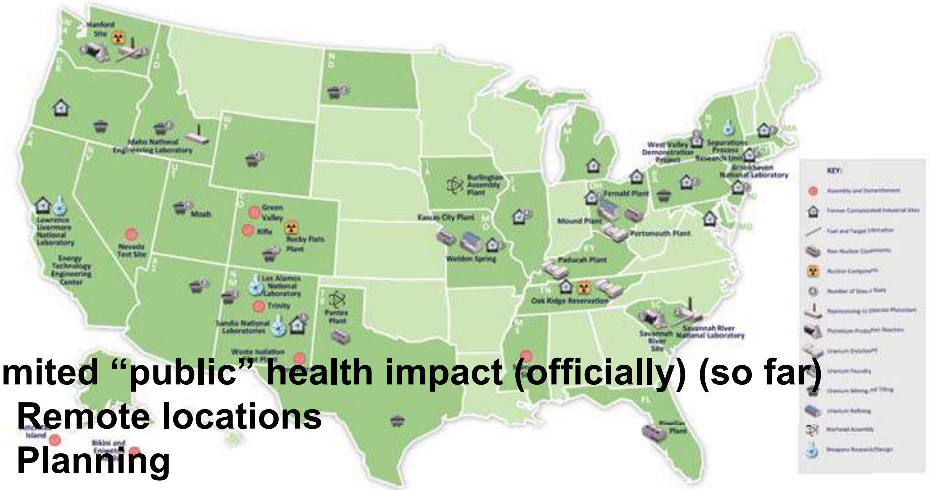
Waste Isolation?



Limited "public" health impact (officially) (so far)

- **Remote locations**
- **Planning**
- **Regulation**

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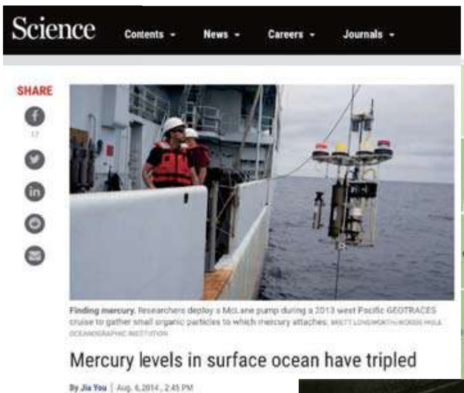
Nuclear Weapon Production Sites: Waste Isolation?



Love Canal

Cuyahoga River

Minamata, Japan

Courtesy to Will Stringfellow

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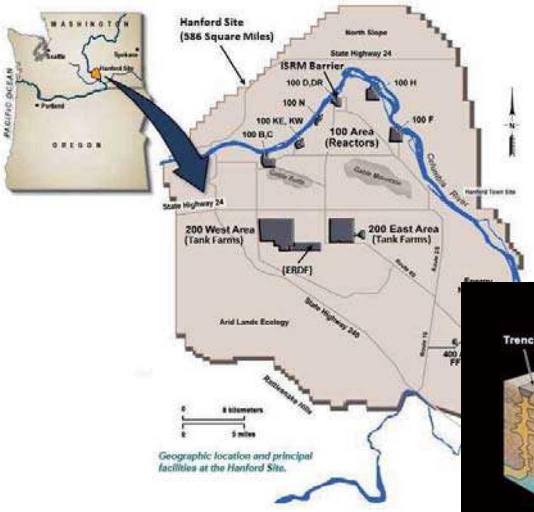
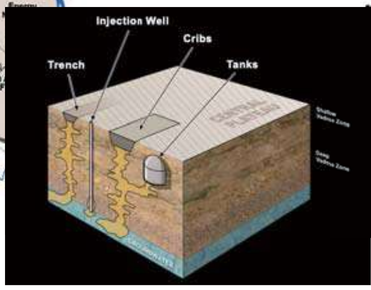
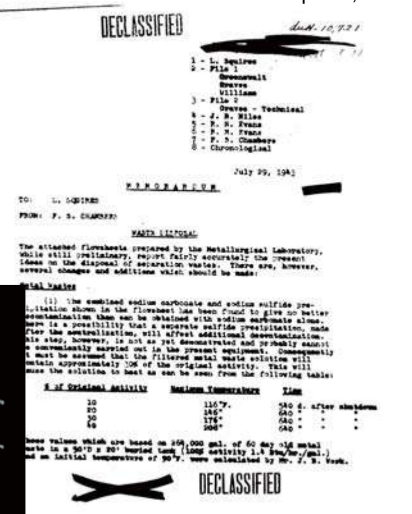
Hanford Site: Thick Vadose Zone





Technical Document on Waste Disposal, 1943



DECLASSIFIED

[redacted]

1 - L. Squires
2 - File 1
Greenewalt
Graves
Williams
3 - File 2
Graves - Technical
4 - J. B. Miles
5 - R. R. Evans
6 - P. M. Evans
7 - F. S. Chambers
8 - Chronological

July 29, 1943

MEMORANDUM [redacted]

TO: L. SQUIRES

FROM: F. S. CHAMBERS

WASTE DISPOSAL

The attached flowsheets prepared by the Metallurgical Laboratory, while still preliminary, report fairly accurately the present ideas on the disposal of separation wastes. There are, however, several changes and additions which should be made:

Metal Wastes

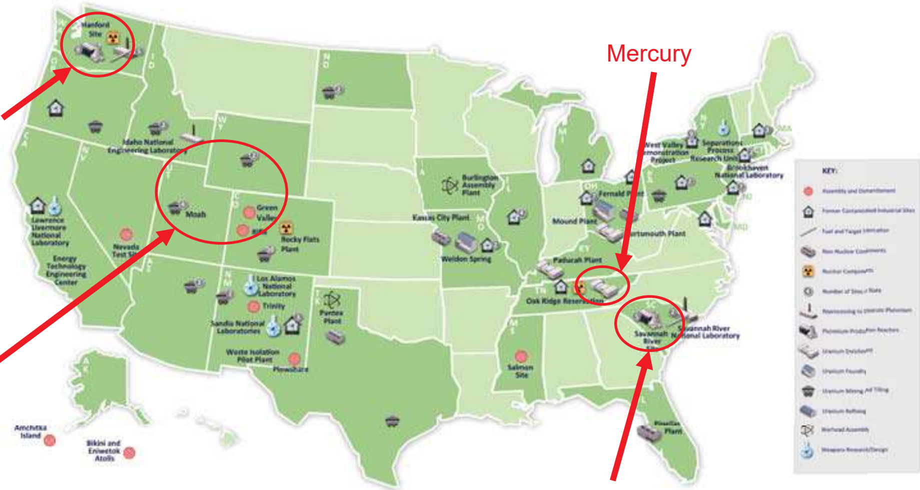
(1) The combined sodium carbonate and sodium sulfide precipitation shown in the flowsheet has been found to give no better decontamination than can be obtained with sodium carbonate alone. There is a possibility that a separate sulfide precipitation, made after the neutralization, will affect additional decontamination. This step, however, is not as yet demonstrated and probably cannot be conveniently carried out in the present equipment. Consequently it must be assumed that the filtered metal waste solution will contain approximately 10% of the original activity. This will cause the solution to heat as can be seen from the following table:

% of Original Activity	Maximum Temperature	Time
10	116°F.	540 d. after shutdown
20	146°	640 " " "
30	176°	640 " " "
40	206°	640 " " "

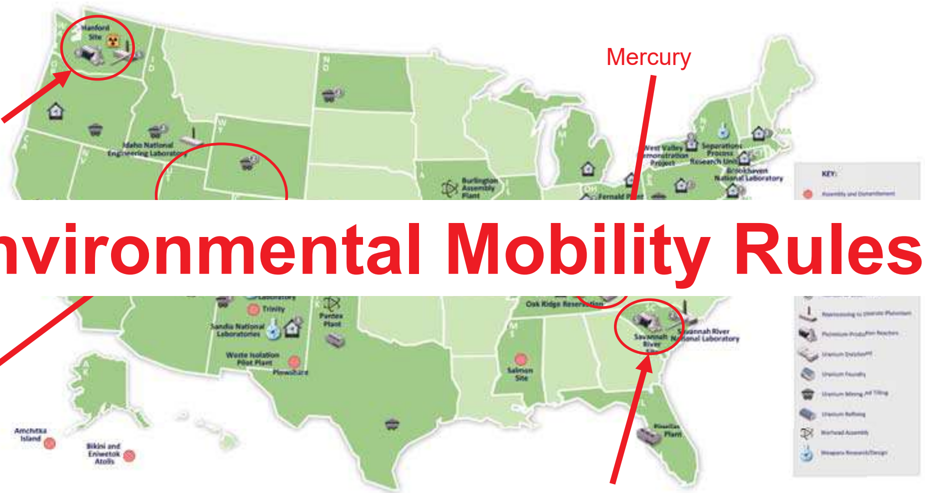
These values which are based on 268,000 gal. of 60 day old metal waste in a 50'D x 20' buried tank (100% activity 1.4 Btu/hr./gal.) and an initial temperature of 90°F. were calculated by Mr. J. B. Work.

[redacted] DECLASSIFIED

Key/Priority Elements

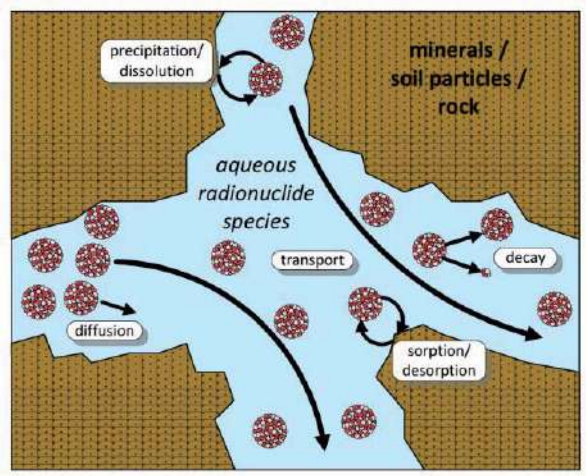


Key/Priority Elements



Environmental Mobility Rules!

Kd: Partition Coefficient



Heuel-Fabianek, 2014

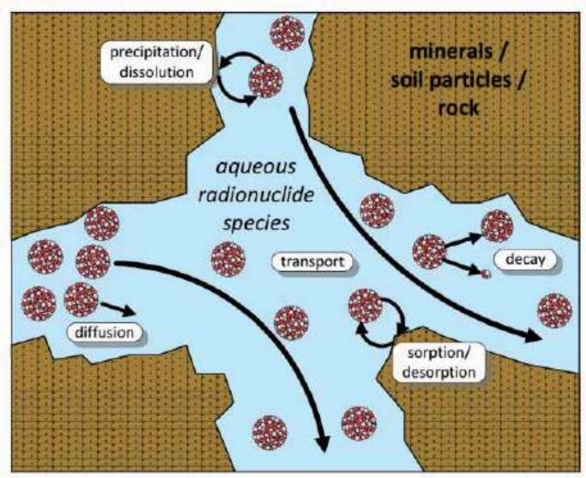
$$K_d = \frac{Solid\ Conc.\,(\frac{mol}{kg})}{Aqueous\ Conc.\,(\frac{mol}{L})}$$

* Assume linear isotherm and equilibrium

Mobility/Velocity of Each Species

$$v = \frac{Groundwater\ Velocity}{1 + \frac{\rho}{\varepsilon} K_d}$$

Kd: Partition Coefficient

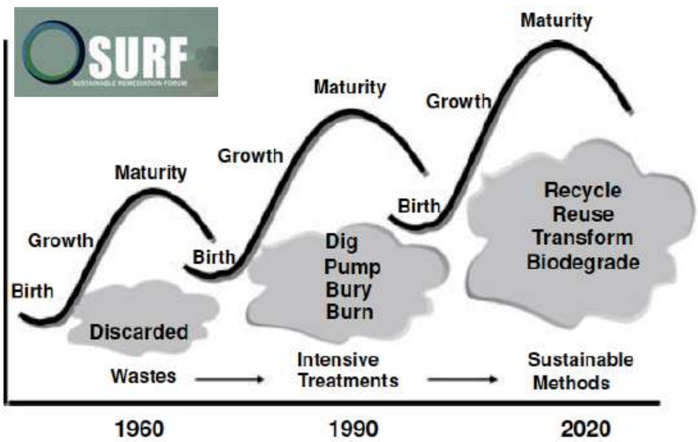


Heuel-Fabianek, 2014

Elements	Kd
H-3	0
I-129	4.5
Tc-99	0.14
Sr-90	20
Uranium	33
Cesium	4.4×10^3
Plutonium	1.2×10^3

Yucca Mountain Assessment, Biosphere Model

Environmental Remediation: Evolution



Sustainable Remediation

Trade offs:

Contaminant removal vs

- Cost
- Waste
- CO2 emission
- Energy Use
- Ecological Impacts
- Noise, Air pollution

Long-term Institutional Control

SURF

- Attractive Re-development Planning
  - **Restrictive use** but **added value**
  - Solar farms, parks, factories
- Longer Institutional Control
  - **Long-term monitoring** will be a major life-cycle cost

Former Reilly Tar & Chemical Corporation Plant

Rocky Flats National Wildlife Refuge

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Importance Of Long-Term Monitoring

THE DENVER POST

OPINION › OPINION COLUMNISTS

Activists ignore the science that says Rocky Flats National Wildlife Refuge is safe

Andy Cross, Denver Post file

A herd of elk walks though a valley at the Rocky Flats National Wildlife Refuge on Sept. 25, 2015.

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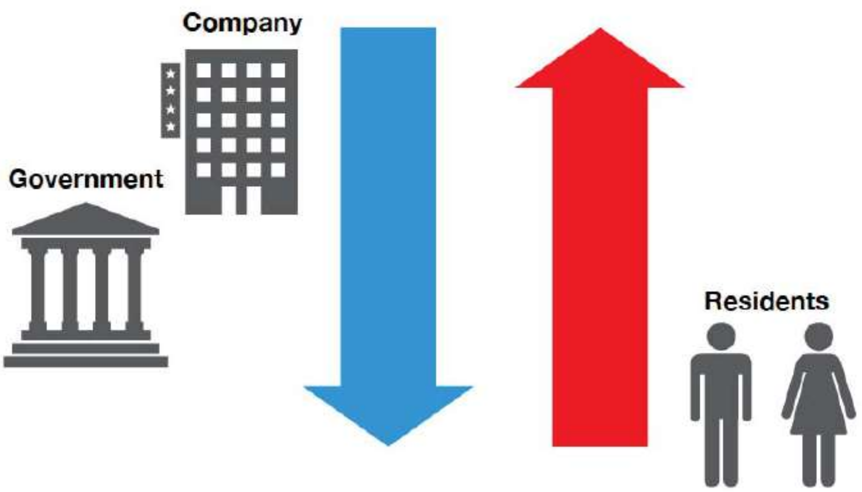
Information Bias @ Environmental Events



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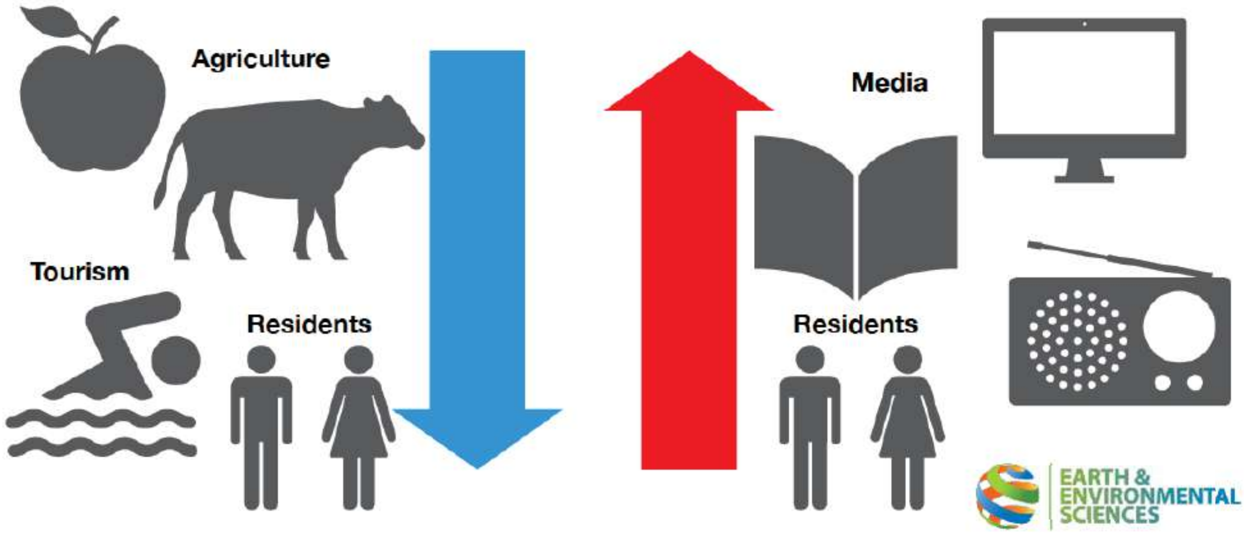
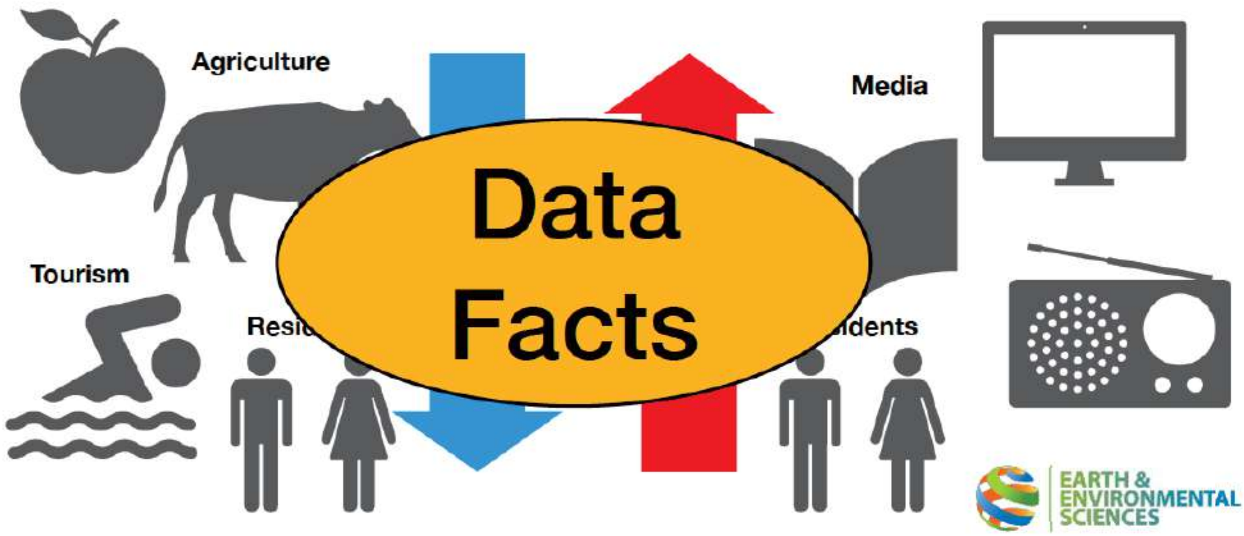
Information Bias @ Environmental Events



Information Bias @ Environmental Events



Two Topics

- Develop Long-term Monitoring Strategies **for Nuclear Contaminated Sites**
  - Fukushima region
  - **Savannah River Site F-Area**

Fukushima Dai-ichi Nuclear Power Plant Accident

March 11, 2011

NYtimes.com

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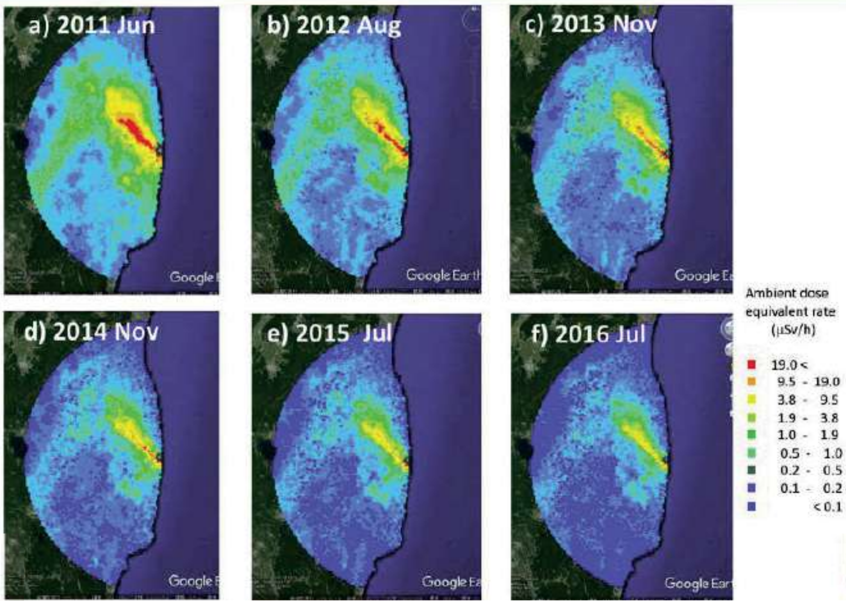
Environmental Recovery



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Challenges in Socioeconomic Recovery

Mirror

NEWS POLITICS SPORT FOOTBALL CELEBS TV MORE

Inside Fukushima ghost town that has laid abandoned since nuclear disaster forced everyone to leave

Thousands of people were forced to flee seven years ago after an earthquake and tsunami caused a nuclear meltdown in Japan

SHARE COMMENTS By Rachel Russell NEWS

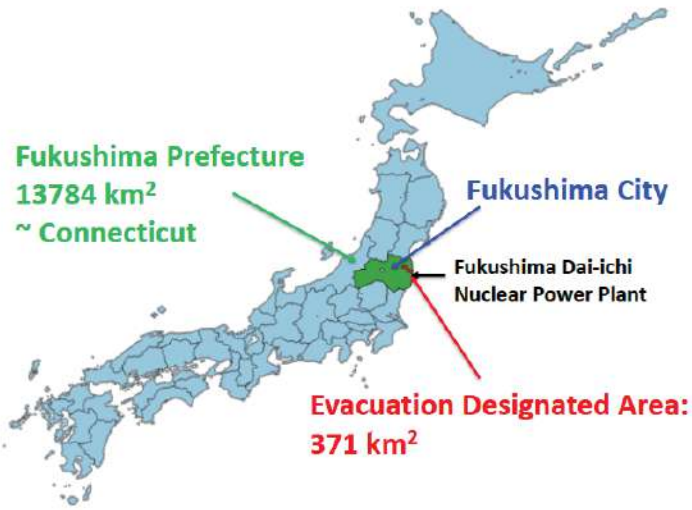
Click for Sound

DISCOVER VIDEO PODCAST NEWSLETTERS BIG THINK

Take hope: This Fukushima disaster map is a fake

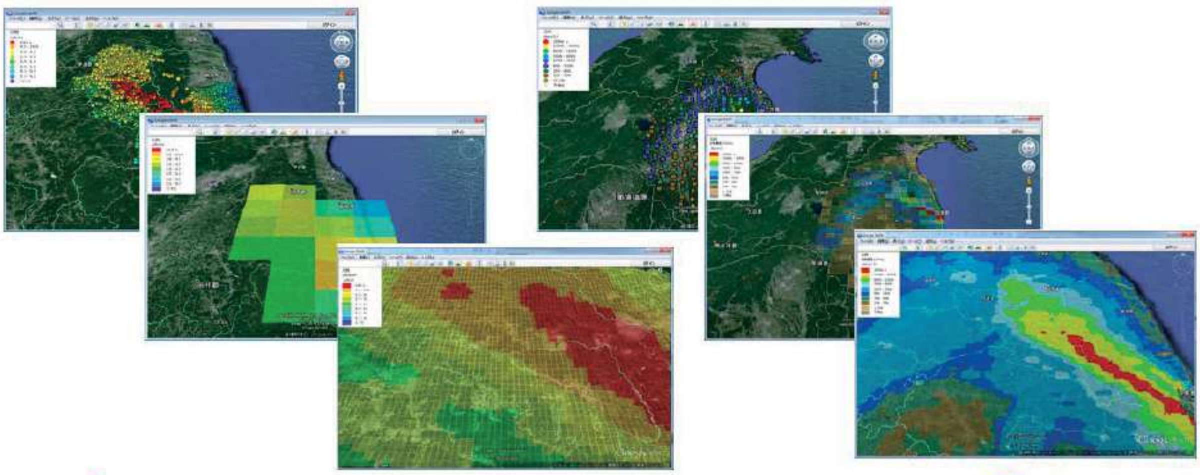
The greatest danger to our planet is not pollution or climate change, but our own despair.

What is Fukushima?

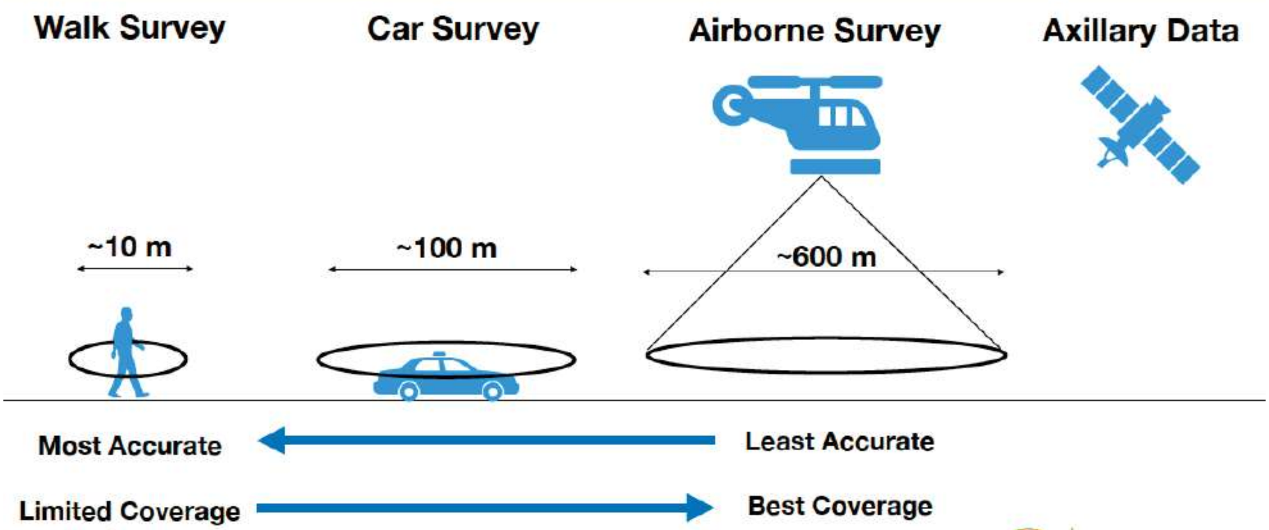


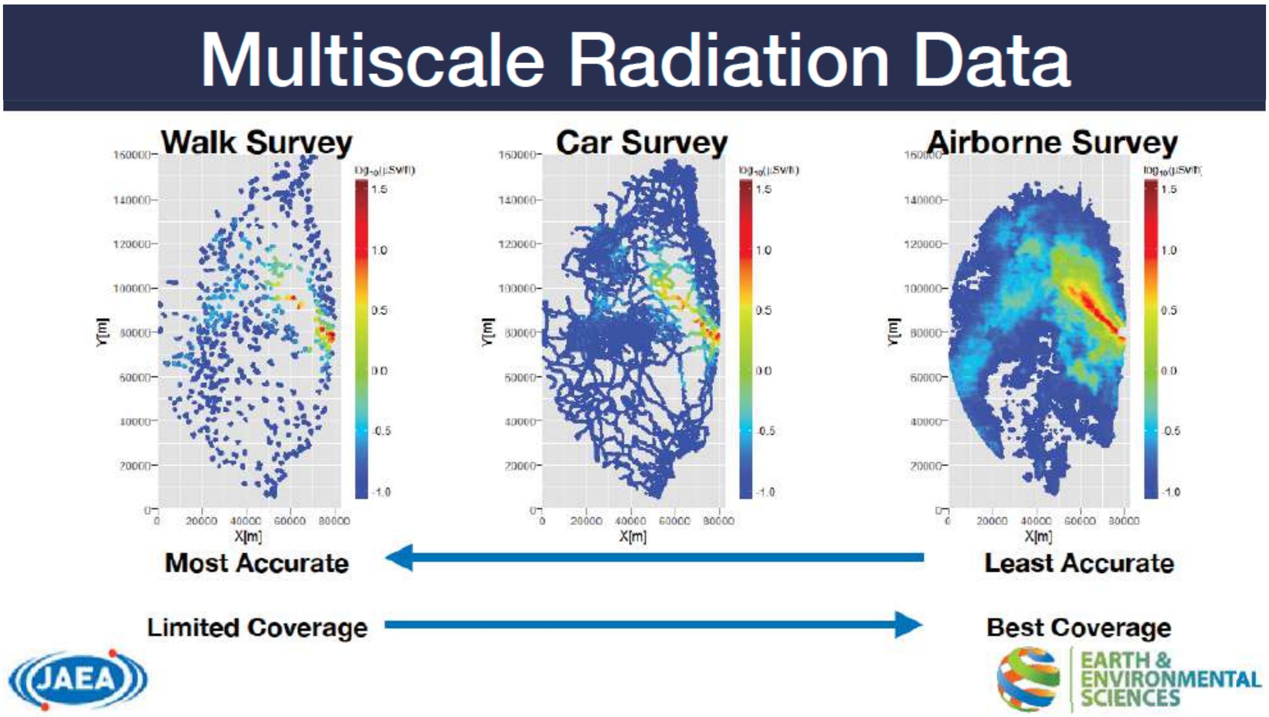
Big Data: Multiscale Multitype Data

Database for Radioactive Substance Monitoring Data (Japan Atomic Energy Agency)



Multiscale Radiation Data Integration





Multiscale Radiation Data
Walk Survey
Car Survey
Airborne Survey
Most Accurate
Least Accurate
Limited Coverage
Best Coverage
JAEA
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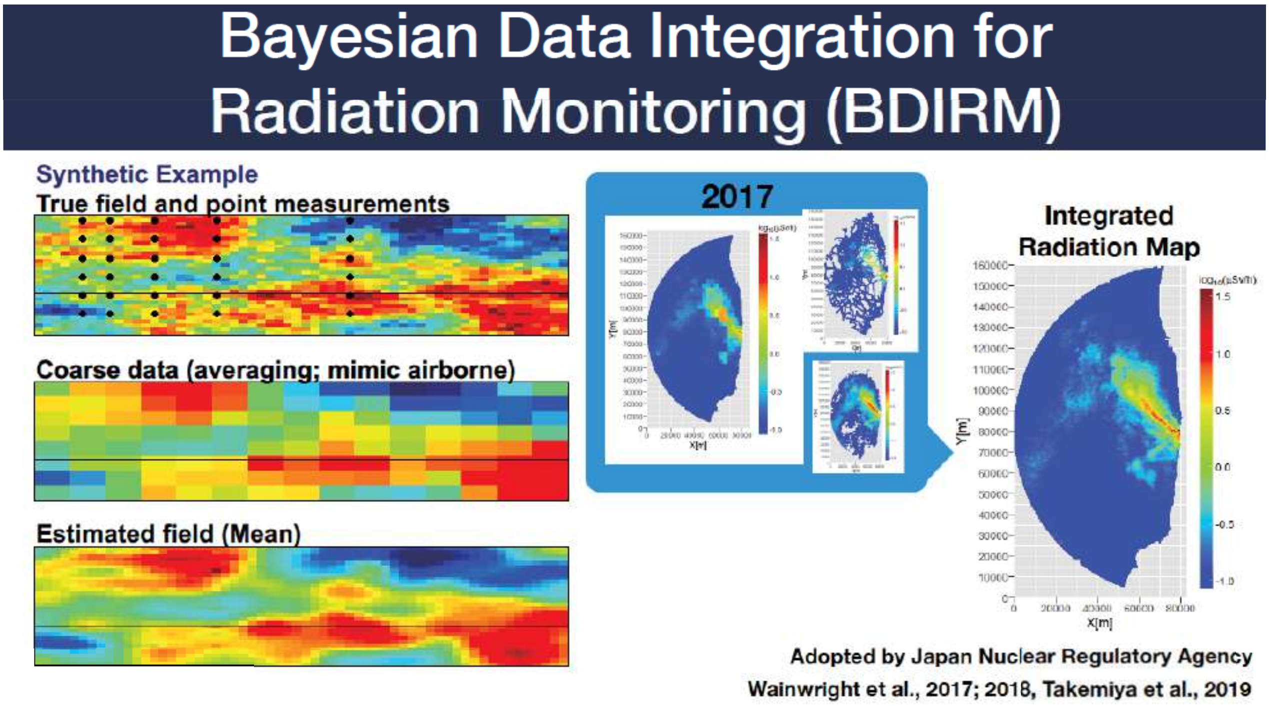
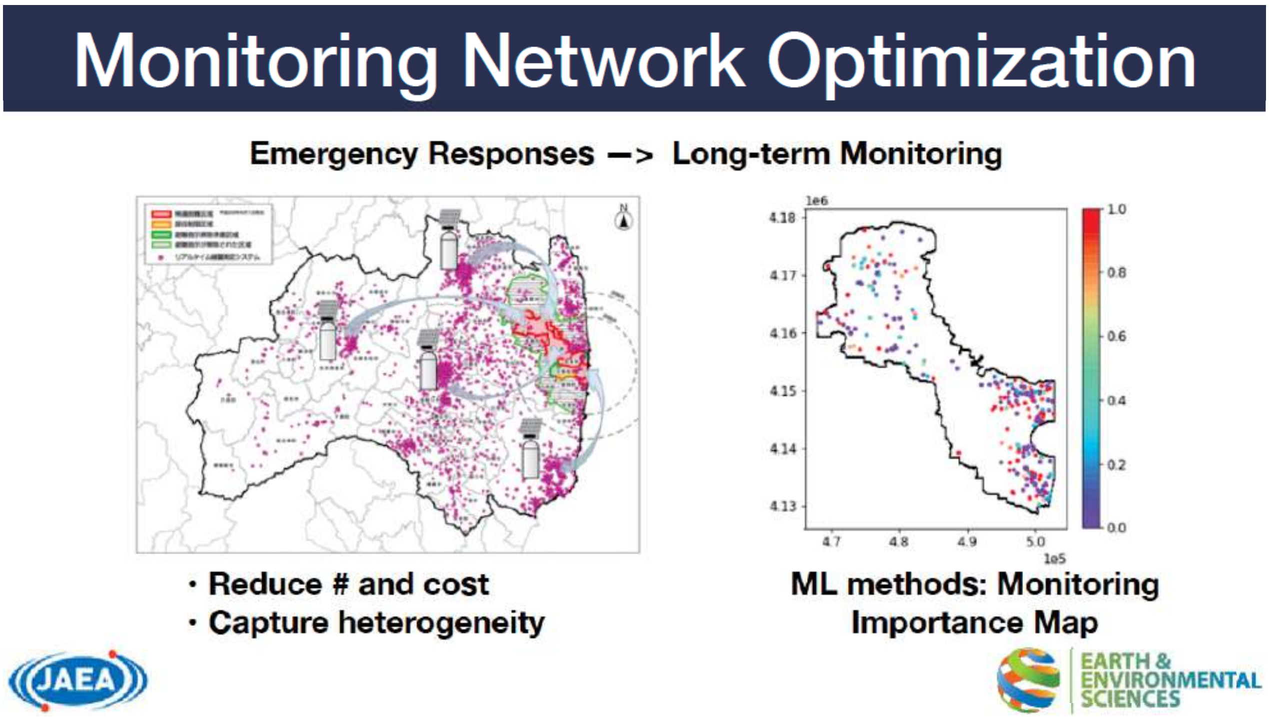


Bayesian Data Integration for Radiation Monitoring (BDIRM)
Synthetic Example
True field and point measurements
Coarse data (averaging; mimic airborne)
Estimated field (Mean)
2017
Integrated Radiation Map
Adopted by Japan Nuclear Regulatory Agency
Wainwright et al., 2017; 2018, Takemiya et al., 2019



Monitoring Network Optimization
Emergency Responses –> Long-term Monitoring
• Reduce # and cost
• Capture heterogeneity
ML methods: Monitoring Importance Map
JAEA
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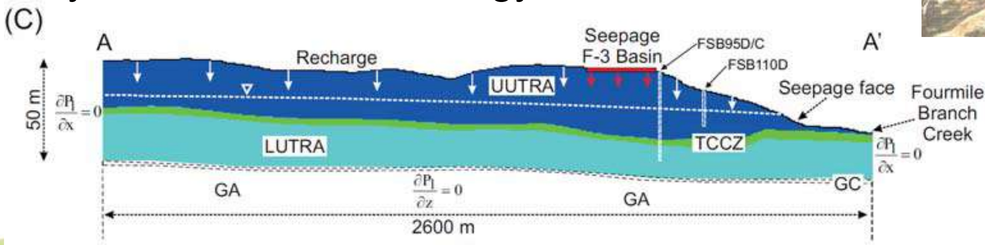
Savannah River Site F-Area

- **Disposal activities**:
  - Disposal of low-level radioactive, acid waste solutions (1955–1989)
  - Acidic plume with radionuclides (pH 3–3.5, U, ^{90}Sr, ^{129}I, ^{99}Tc, ^{3}H)
- Many datasets → Technology Test Bed



Bea et al., 2013

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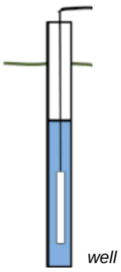
Current Groundwater Monitoring

- **Groundwater Sampling → Mass Spec**
  - 10s – 100s of wells
  - Contamination issues (requires training, equipment)
  - Temporally sparse: every quarterly, annually → Miss anomalies
  - Compliance only (no analytics)



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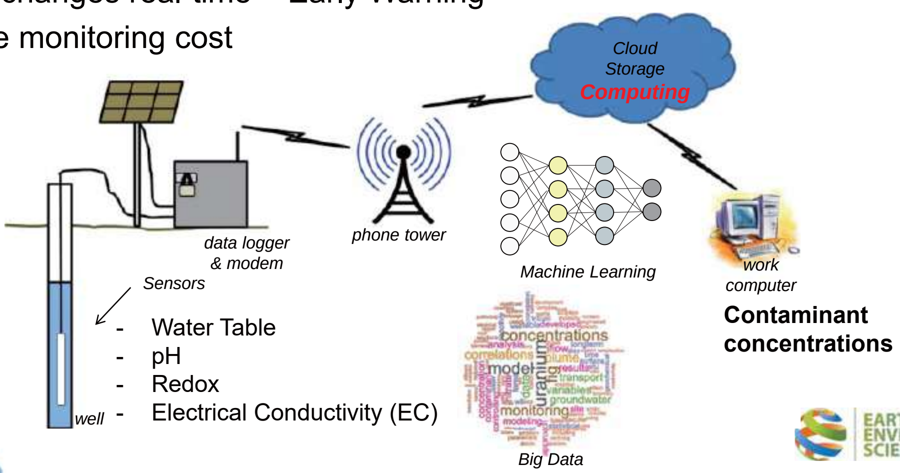
New Paradigm of Long-Term Monitoring

- **Low-cost in situ sensors, wireless network, cloud computing**
  - → Autonomous continuous monitoring
  - → Detect changes real-time = Early Warning
  - → Reduce monitoring cost



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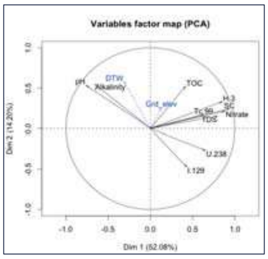
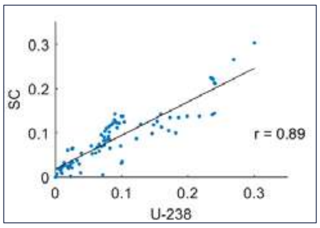
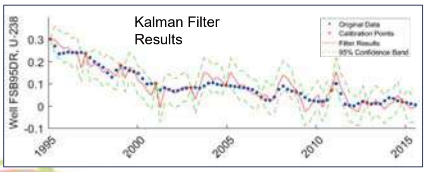
Data Analytics Workflow



Exploratory Data Analysis

Quantification of Correlations

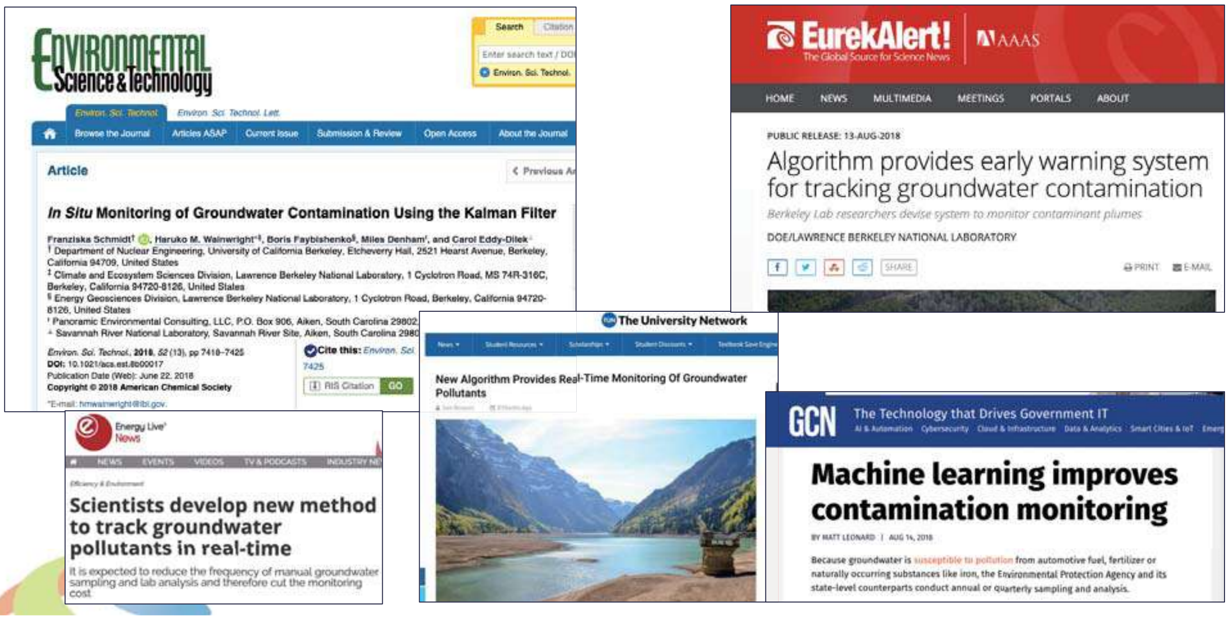




Contaminant Concentration Estimation

Machine Learning

Big Interest in ML x Environment

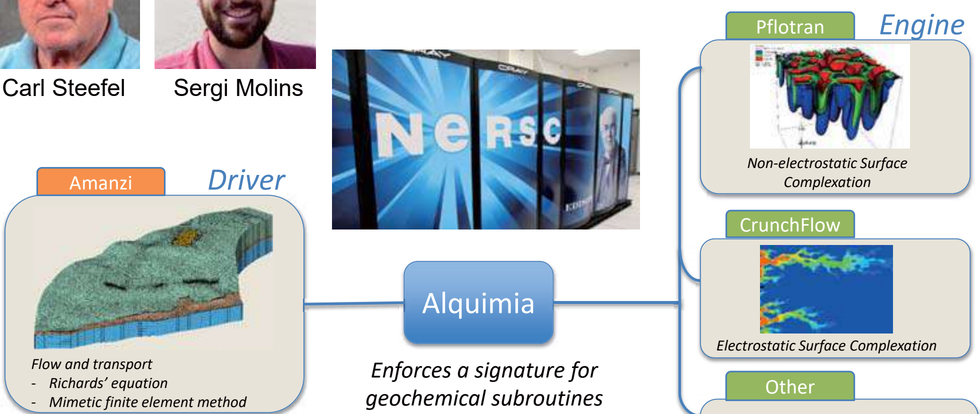


Reactive Transport Modeling: Amanzi

David Moulton

Carl Steefel

Sergi Molins

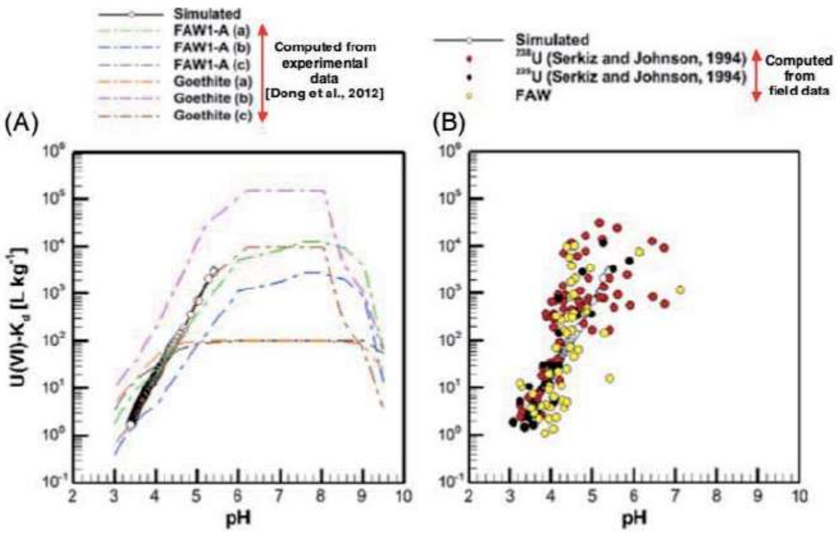


SRS F-Area: Geochemical Model

Reaction	log K (25 °C)
Aqueous species[1]	
$(UO_2)_2(OH)_2^{+2} \leftrightarrow 2UO_2^{+2} + 2H_2O - 2H^+$	5.62
$(UO_2)_2CO_3(OH)_3^- \leftrightarrow 2UO_2^{+2} + 3H_2O + HCO_3^- - 4H^+$	11.18
$(UO_2)_2OH^{+3} \leftrightarrow 2UO_2^{+2} + H_2O - H^+$	2.7
$(UO_2)_3(CO_3)_6^{-6} \leftrightarrow 3UO_2^{+2} + 6HCO_3^- - 6H^+$	7.97
$(UO_2)_3(OH)_4^{+2} \leftrightarrow 3UO_2^{+2} + 4H_2O - 4H^+$	11.9
$UO_2(OH)_4^{-2} \leftrightarrow UO_2^{+2} + 4H_2O - 4H^+$	32.4
$(UO_2)_3(OH)_5^+ \leftrightarrow 3UO_2^{+2} + 5H_2O - 5H^+$	15.55
$(UO_2)_3(OH)_7^- \leftrightarrow 3UO_2^{+2} + 7H_2O - 7H^+$	32.2
$(UO_2)_3O(OH)_2(HCO_3)^+ \leftrightarrow 3UO_2^{+2} + 3H_2O + HCO_3^- -$	9.68
$(UO_2)_4(OH)_7^+ \rightarrow 4UO_2^{+2} + 7H_2O - 7H^+$	21.9
$UO_2NO_3^+ \leftrightarrow UO_2^{+2} + NO_3^-$	−0.3
$UO_2(OH)^+ \leftrightarrow UO_2^{+2} + H_2O$	5.25
$UO_2(OH)_2(aq) \leftrightarrow UO_2^{+2} + 2H_2O - 2H^+$	12.15
$UO_2(OH)_3^- \leftrightarrow UO_2^{+2} + 3H_2O - 3H^+$	20.25
$UO_2CO_3(aq) \leftrightarrow UO_2^{+2} + HCO_3^- - H^+$	0.39
$UO_2(CO_3)_2^{-2} \leftrightarrow UO_2^{+2} + 2HCO_3^- - 2H^+$	4.05
$UO_2(CO_3)_3^{-4} \rightarrow UO_2^{+2} + 3HCO_3^- - 3H^+$	9.14
$CaUO_2(CO_3)_3^{-2} \rightarrow Ca^{+2} + UO_2^{+2} + 3HCO_3^- - 3H^+$	3.8
$Ca_2UO_2(CO_3)_3(aq) \leftrightarrow 2Ca^{+2} + UO_2^{+2} + 3HCO_3^- - 3H^+$	0.29
$MgUO_2(CO_3)_3^{-2} \leftrightarrow Mg^{+2} + UO_2^{+2} + 3HCO_3^- - 3H^+$	5.19
$UO_2SiO(OH)_3^+ \leftrightarrow SiO_2(aq) + UO_2^{+2} + 2H_2O - H^+$	2.48
Surface and exchange species[2]	
$(>k\text{-}OH)_2UO_2^+ \leftrightarrow 2 >k\text{-}OH^{-0.5} + UO_2^{+2}$	−5.3
$(>k\text{-}OH)_2UO_2CO_3^- \leftrightarrow 2 >k\text{-}OH^{-0.5} + UO_2^{+2} + HCO_3^- - H^+$	−6.2
$>k\text{-}OH_2^{+0.5} \leftrightarrow >k\text{-}OH^{-0.5} + H^+$	−4.9
$>k\text{-}OHNa^{+0.5} \rightarrow >k\text{-}OH^{-0.5} + Na^+$	2.1
$>k\text{-}OH_2NO_3^{-0.5} \leftrightarrow >k\text{-}OH^{-0.5} + H^+ + NO_3^-$	−4.9
$>k_2UO_2 \leftrightarrow 2 >k^- + UO_2^{+2}$	−7.1
$>kNa \leftrightarrow >k^- + Na^+$	−2.9
$>kH \leftrightarrow >k^- + H^+$	−4.5
$>k_2Ca \rightarrow 2 >k^- + Ca^{+2}$	−6.8
$>k_3Al \rightarrow 3 >k^- + Al^{+3}$	−8
$(>Fe\text{-}OH)_2UO_2^+ \leftrightarrow 2 >Fe\text{-}OH^{-0.5} + UO_2^{+2}$	−14.11
$(>Fe\text{-}OH)_2UO_2CO_3^- \leftrightarrow 2 >Fe\text{-}OH^{-0.5} + UO_2^{+2} + HCO_3^- - H$	−4.35
$>Fe\text{-}OH_2^{+0.5} \leftrightarrow >Fe\text{-}OH^{-0.5} + H^+$	−9.18
$(>Fe\text{-}OH)_2CO_2^- \leftrightarrow 2 >Fe\text{-}OH^{-0.5} + H^+ + HCO_3^- - 2H_2O$	−12.23
$>Fe\text{-}OCO_2Na^{-0.5} \leftrightarrow >Fe\text{-}OH^{-0.5} + Na^+ + HCO_3^- - H_2O$	−3.28
$>qz\text{-}OH_2^+ \leftrightarrow >qz\text{-}OH + H^+$	1.1[3]
$>qz\text{-}O^- \leftrightarrow >qz\text{-}OH - H^+$	8.1[3]
$>qz\text{-}ONa \leftrightarrow >qz\text{-}OH - H^+ + Na^+$	6.8[4]

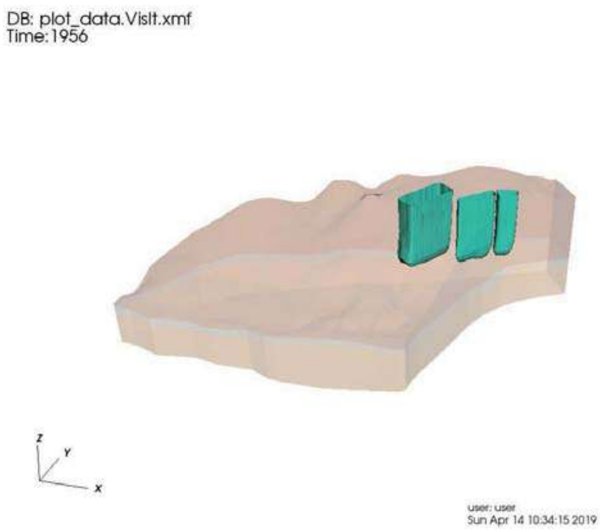
Arora et al, 2017

SRS F-Area: Surface Complexation Model



Dong et al., 2012; Bea et al, 2013

3D Uranium Plume Evolution

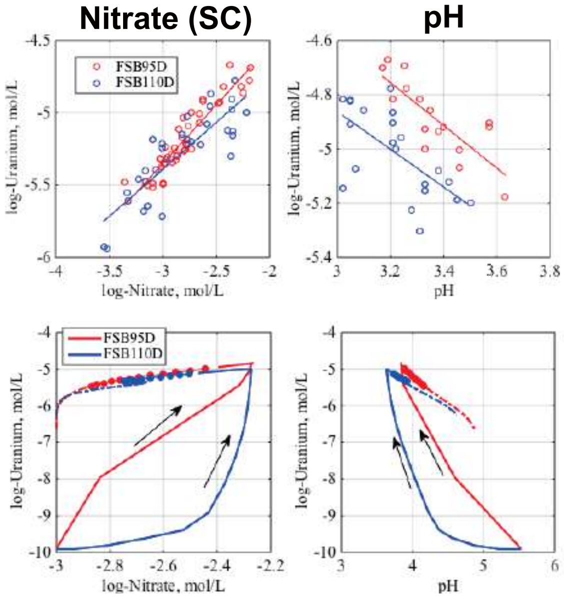


Uranium Plume:
Residual contaminants
- Under the basins
- Within Tan Clay

In situ Variables vs Contaminant Concentrations

Measured

Simulated



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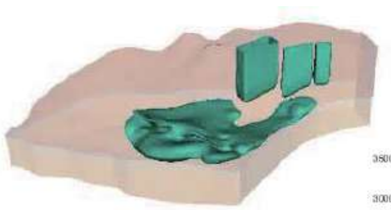
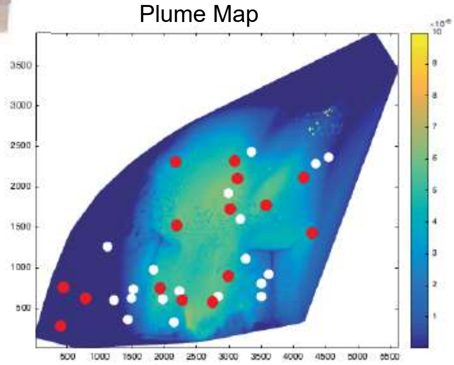
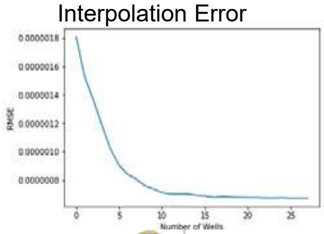
Monitoring Optimization

3D Simulation Result





- Reduce # wells effectively
- Capture the spatial heterogeneity of the plume with minimal # wells



Algorithm

- Gaussian Process Models
- Greedy Algorithm

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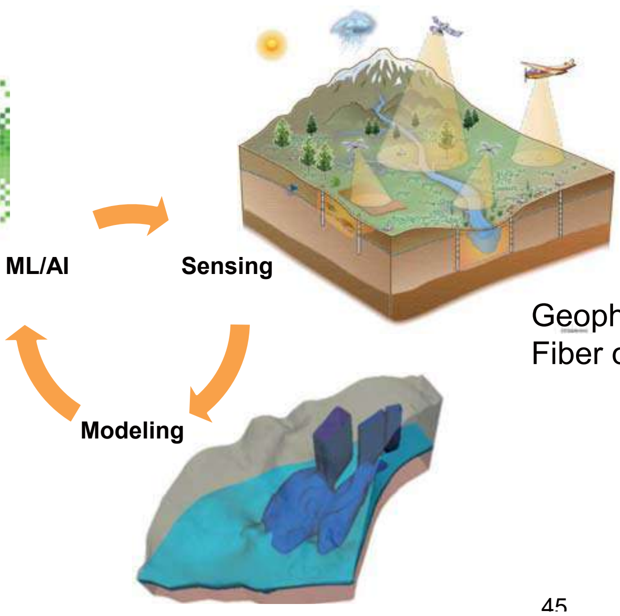
Long-term Monitoring: Next Step

AI FOR SCIENCE TOWN HALL



Remote sensing

- Wetland
- Surface Barrier
- ET

Geophysics
Fiber optics

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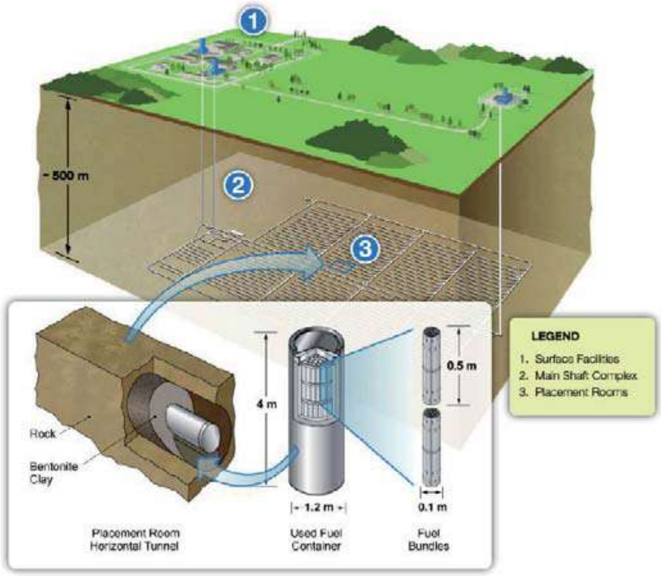
Nuclear Waste DisposalSite

Engineered barrier system:
- Canister
- Buffer (Clay/Bentonite)

Natural barrier system:
- Deep subsurface
- Host rock (low permeability rock)



Summary

- **What can we learn from existing nuclear contamination for nuclear waste?**
  - Environmental mobility of radionuclides is critical
  - Engineering solutions for isolation could be possible
    - Nuclear sectors: more awareness, tighter regulations
  - Net environmental impact or broader environmental perspective is needed
    - Rads vs others; contaminant removal vs side effects
  - Long-term monitoring is important for public assurance
- **New paradigm for long-term monitoring based on new sensors/ML/AI**
  – Multiscale data integration and monitoring placement optimization for long-term monitoring: → B-DRAM and machine learning
  – Real-time monitoring of groundwater contamination
  – Public assurance, early warning, cost reduction

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Thank You!

Contact
Haruko Wainwright
HMWainwright@LBL.gov
* Summer internship positions and exchange programs at LBNL

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DOE Office of Science
Japan Atomic Energy Agency

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