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Radioiodine/iodine Attenuation Mechanisms in Hanford Groundwater

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Waste from Plutonium Production at Hanford

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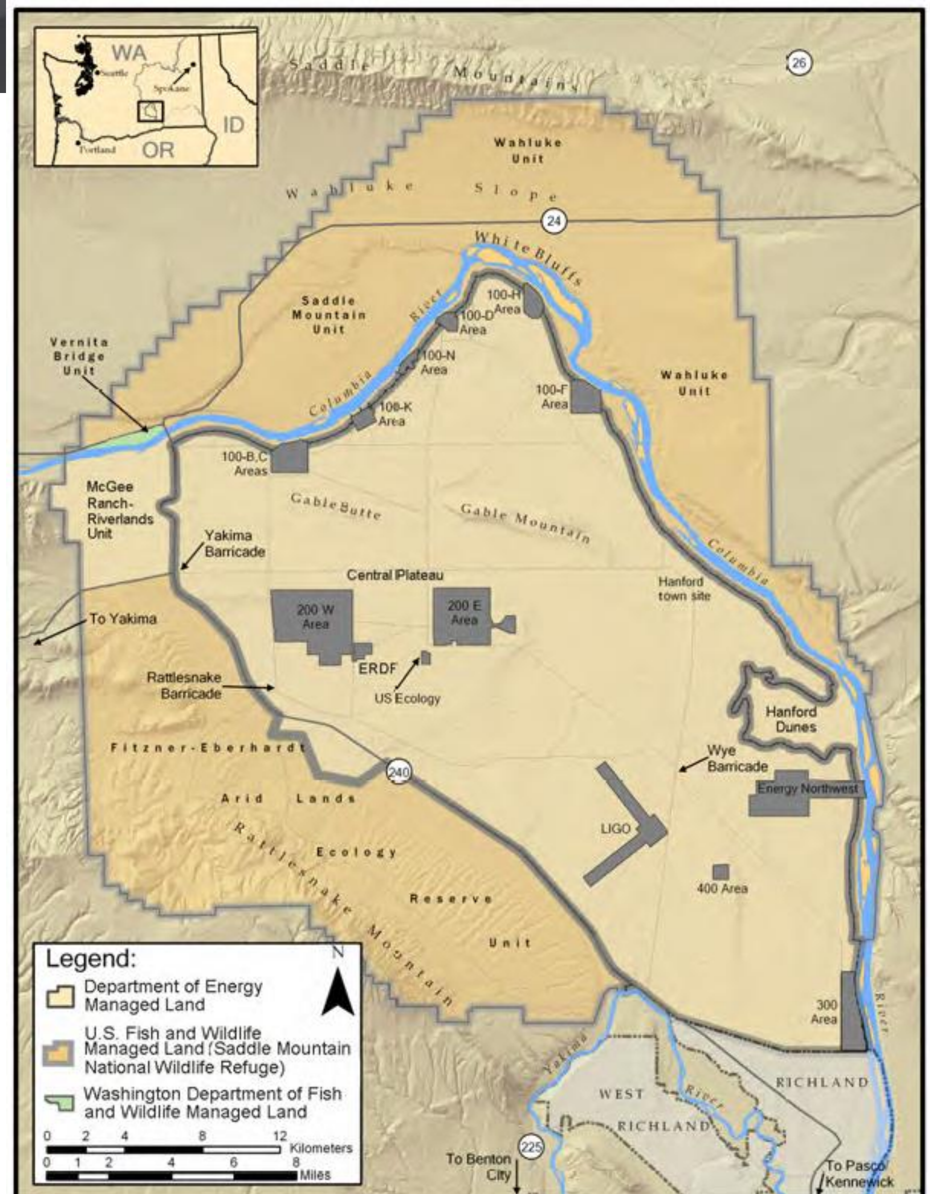
- 586 sq. miles
  - Shrub steppe desert in southeast WA
- Production period from 1944 to 1987
- 110,000 tons of nuclear fuel was processed
- Billions of gallons of liquid waste produced
  - Stored in single-shell and double-shell tanks
  - Discharged to liquid disposal sites (e.g., pits, cribs and trenches)



A Global Perspective of Radioiodine in Environment

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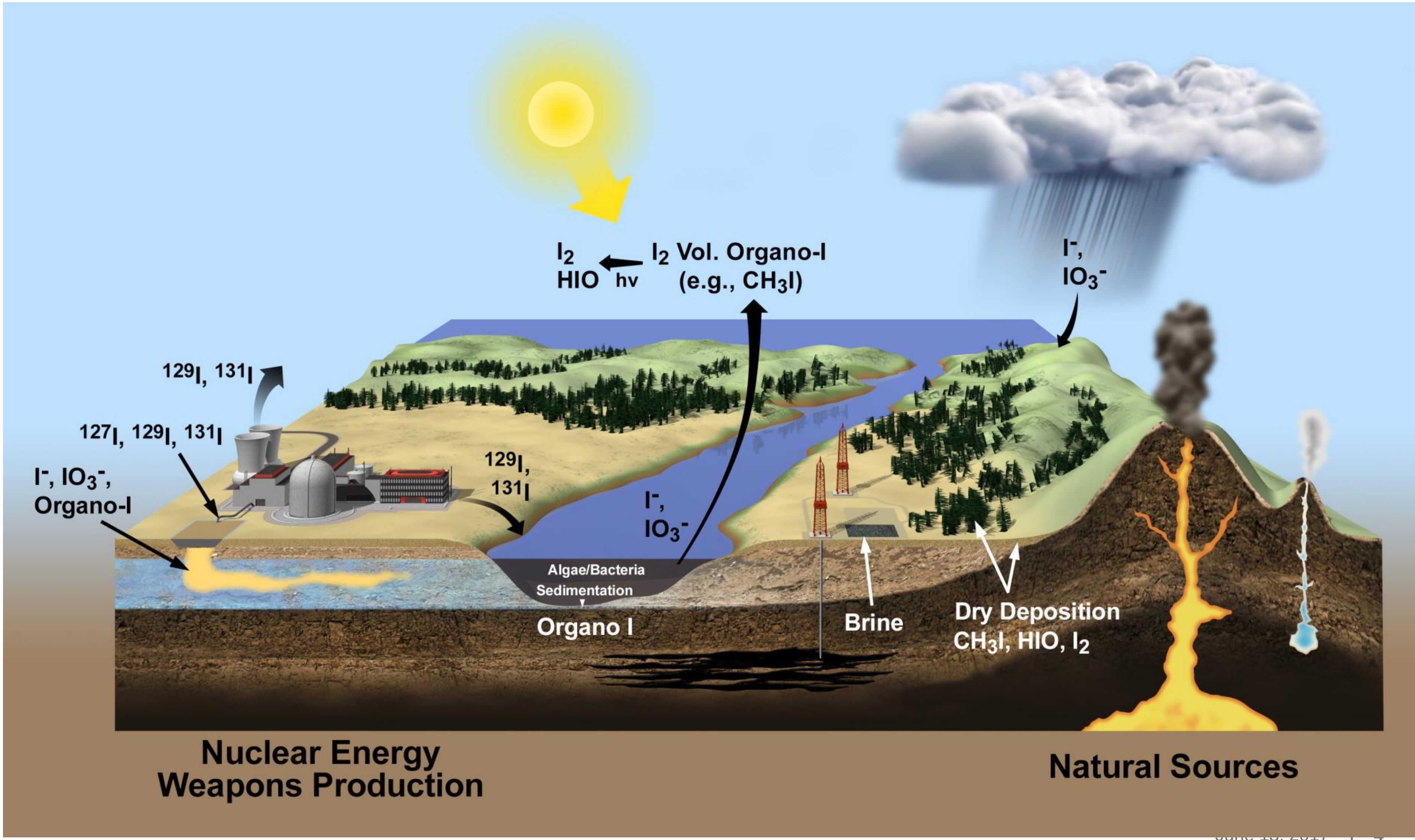
Source	^{129}I Mass Released (kg)
Fuel reprocessing at La Hague (France)	3800 [a]
Fuel reprocessing Sellafield (UK)	1400 [a]
Hanford Site	266 [b]
Natural hydrosphere and atmosphere	100 [c]
Atmospheric weapons testing	50 [b]
Savannah River Site	32 [d]
Nevada Test Site underground nuclear testing	10 [b]
Chernobyl	1-2 [b]
Fukushima	1.2 [e]

(a) Hou et al. 2009
(b) Raisbeck and Yiou 1999
(c) Bustad et al. 1983
(d) Kantelo et al. 1990
(e) Hou et al. 2013

Global Iodine Cycle



Iodine Inventory in Hanford Waste

I-129 Inventory Category	Estimate	Discussion and References
Total generated by production reactors	49.4 Ci	Based on calculation using the 2002 ORIGIN2 fuel activity estimate (Watrous et al. 2002). This estimate is well known and based on fuel irradiation histories.
Stored in single-shell and double-shell tanks	29.0 Ci [(a)]	Best Basis Inventory (BBI) obtained from the Tank Waste Information Network System (April 23, 2015) (https://twins.labworks.org/twinsdata/default.htm). Significant uncertainty remains with this estimate.
Discharged to liquid disposal sites	4.7 Ci	From Hanford's Soil Inventory Model (Corbin et al. 2005). Uncertainty estimates were developed for individual waste sites that ranged from 20% to almost 400%.
Released to the atmosphere	Unknown	Estimates of magnitude of these potential releases are not available. This remains one of the main uncertainties limiting development of a true mass balance for Hanford ^{129}I.
Captured by offgas absorbent devices	Unknown	Devices known as "silver reactors" were used to capture iodine at chemical separations plants (PUREX, B-Plant, T-Plant, and REDOX). The ^{129}I inventory captured in this manner is not known. Some of these devices remain at the canyon facilities and some are in solid waste burial grounds.

(a) The BBI underwent a significant update in 2004 (Higley et al. 2004), which reduced the tank inventory estimate from 48.2 to 31.8 Ci based on improved models of separations processes. This change removed the previous conservative assumption that essentially all of the ^{129}I sent to the separations plants exited those plants in waste streams sent to tank farms. Subsequent revisions to the BBI have replaced generic estimates for specific waste streams with sample-based estimates from the tanks.

Iodine Inventory and Flux to Groundwater

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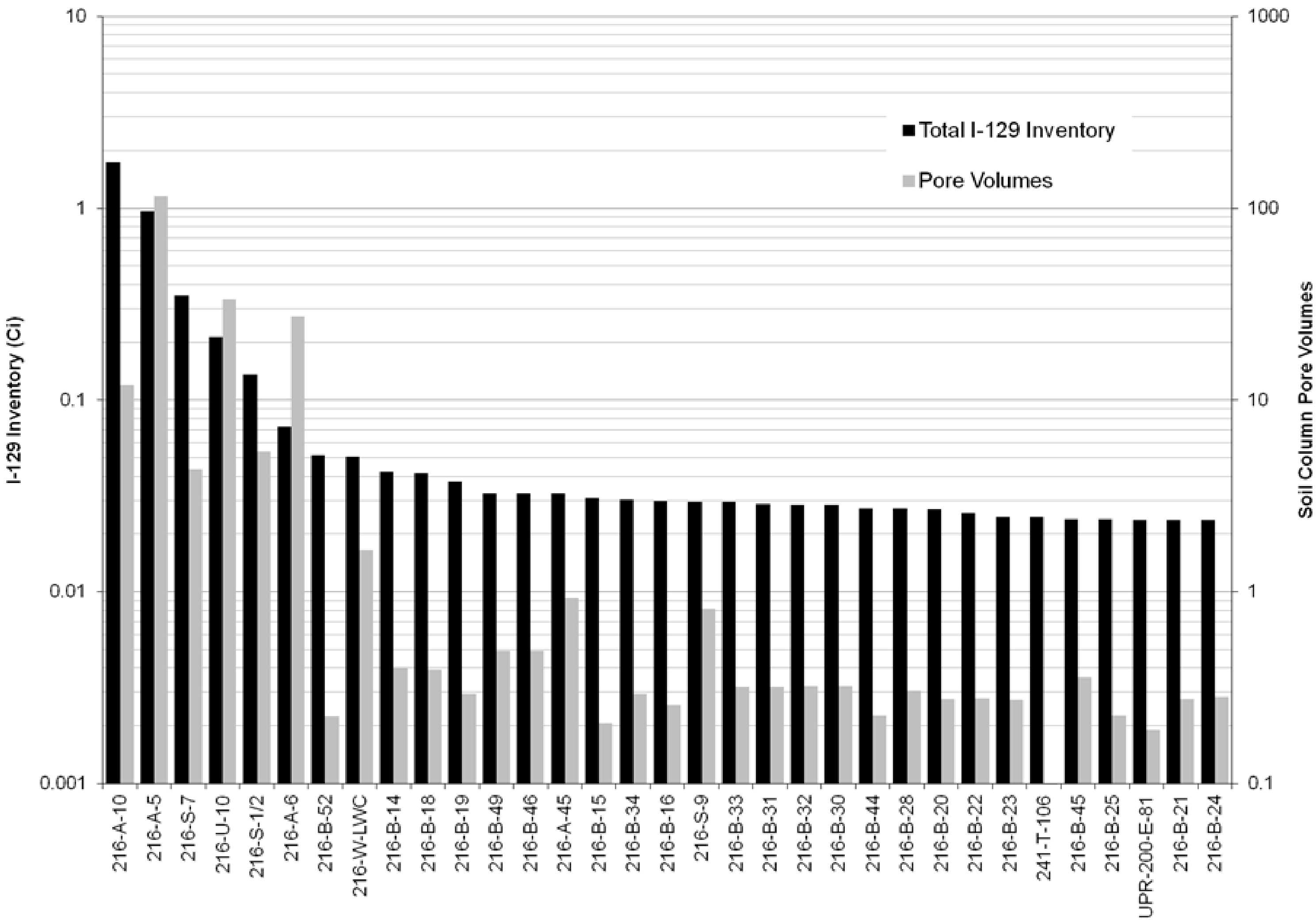
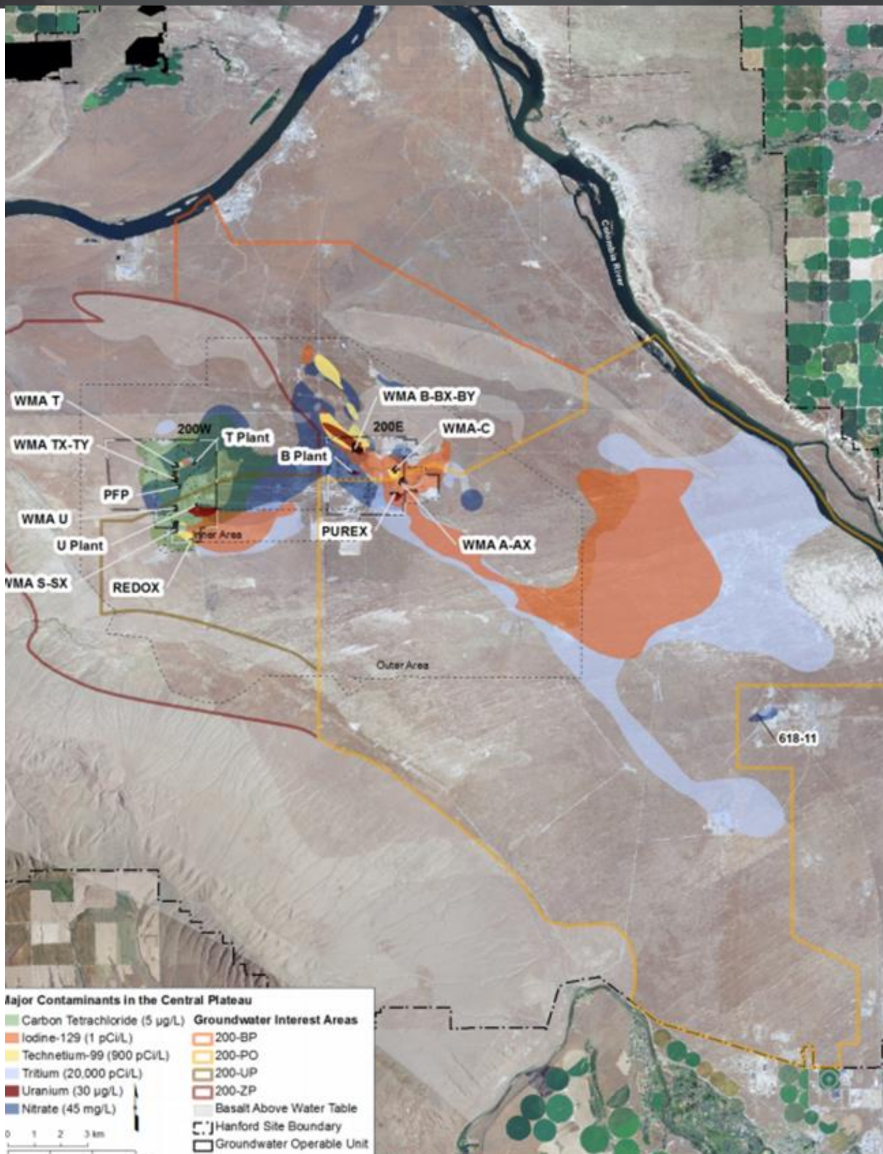


Iodine-129 Contamination at Hanford

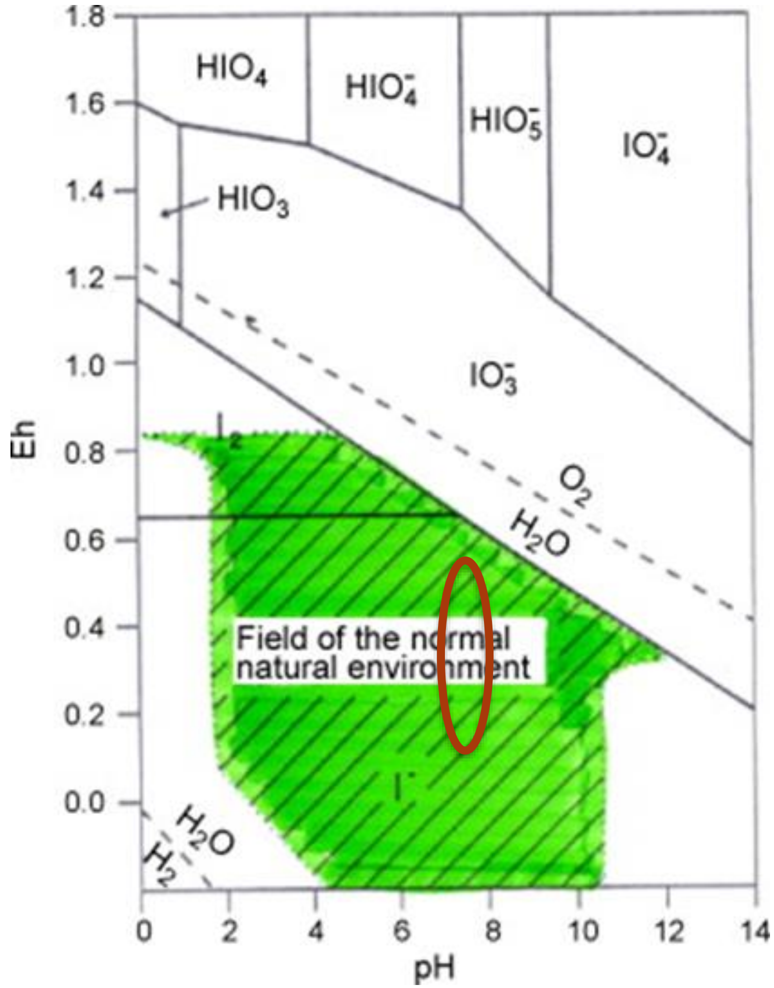


- ^{129}I is found in two separate plumes in the 200 Area of Hanford Site
- These plumes cover >50 km^2; ~3.5 pCi/L (DWS: 1 pCi/L)
- ^{127}I concentrations are approximately 200 times higher than ^{129}I
- Hydraulic containment is the current remedial action
- Treatment technologies are unavailable; are complicated by the geochemistry (alkaline, oxygenic) of groundwater at Hanford site; ^{127}I competes for reactants added for remediation

Current Conditions Related to Iodine Speciation at Hanford

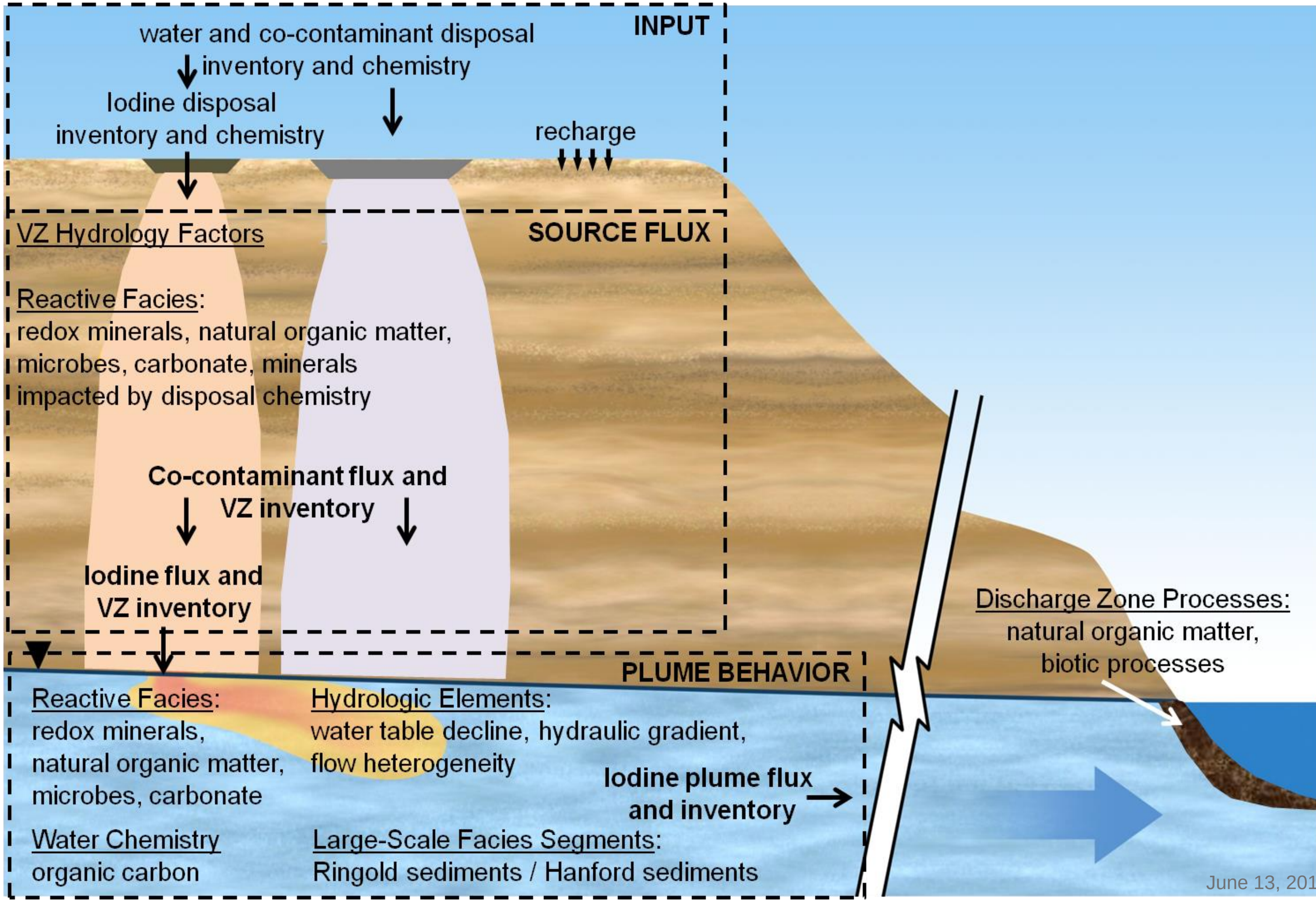
- Speciation at 200 Area:
  - Iodate (IO_3^-) is the prevalent form of iodine, 70.6%
  - Iodide (I^-) , 3.6%
  - Organo-iodine, 25.8%

- ***Speciation is significant because based on chemical thermodynamics, the dominant species should be iodide***

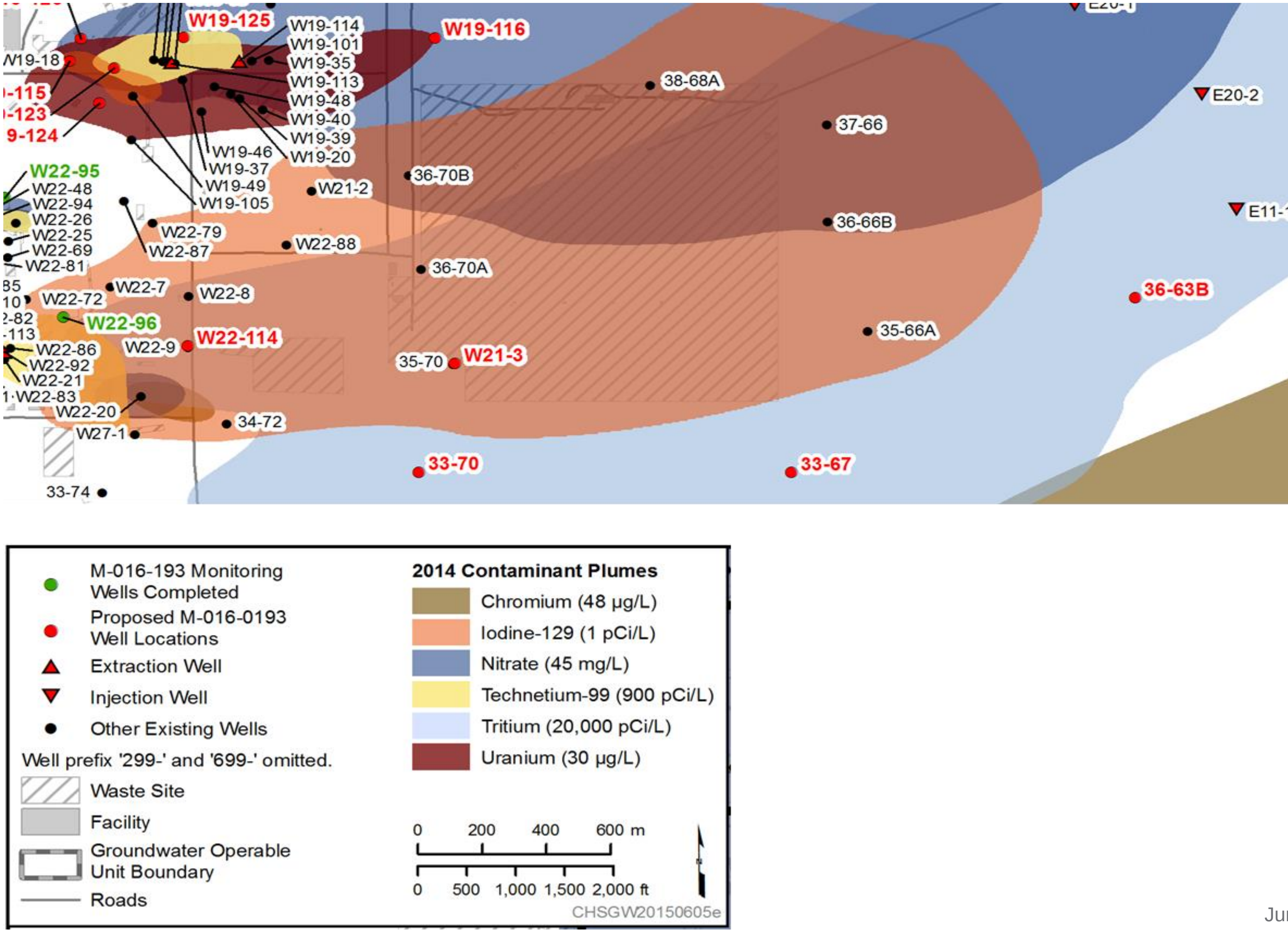


From: Hou et al. 2009

Conceptual Model



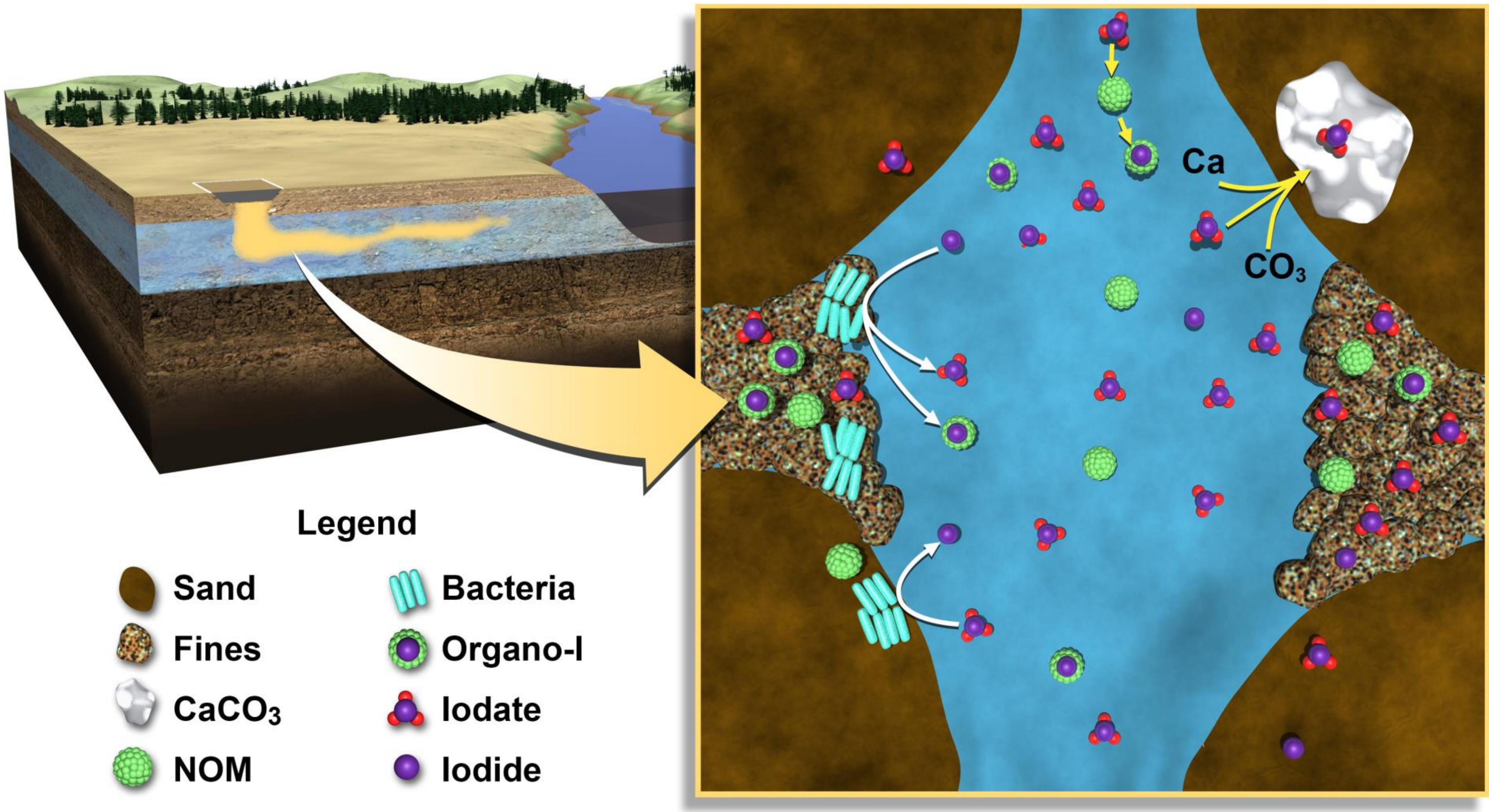
Hanford 200-UP-1 Iodine Plume



Iodine Speciation in Core of Plume

Depth (ft bls)	Depth (ft bwt)	I-129 (pCi/L)	Iodate (µg/L)	Iodide (µg/L)	Nitrate (mg/L)
257.5	7.59	4.01	2.81	3.55	79.7
284	34.09	1.73	17.8	ND	5.31
317.2	67.29	1.49	14.5	ND	4.38
347.6	97.69	ND	17.1	ND	3.5
377.6	127.69	ND	17.6	ND	3.9
427.5	177.59	ND	21.8	ND	6.2
447.2	197.29	ND	22.4	ND	7.53

Biogeochemical Processes Controlling Fate and Transport of Iodine in Hanford Groundwater



Iodine Geochemistry - Precipitation

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- Only small fraction of total iodine was exchangeable with KCl
  - 0.4 to 6.6%
- Low levels of iodine associated with Fe or Mn Oxides
  - 0 to 1.2%
- Major fraction of iodine was strongly bound to sediments
  - Incorporated into $CaCO_3$ - 2.9 to 39.4%
  - Residual fraction (organic carbon) - 57.1 to 90.6%

Iodine Geochemistry – Precipitation (Cont.)

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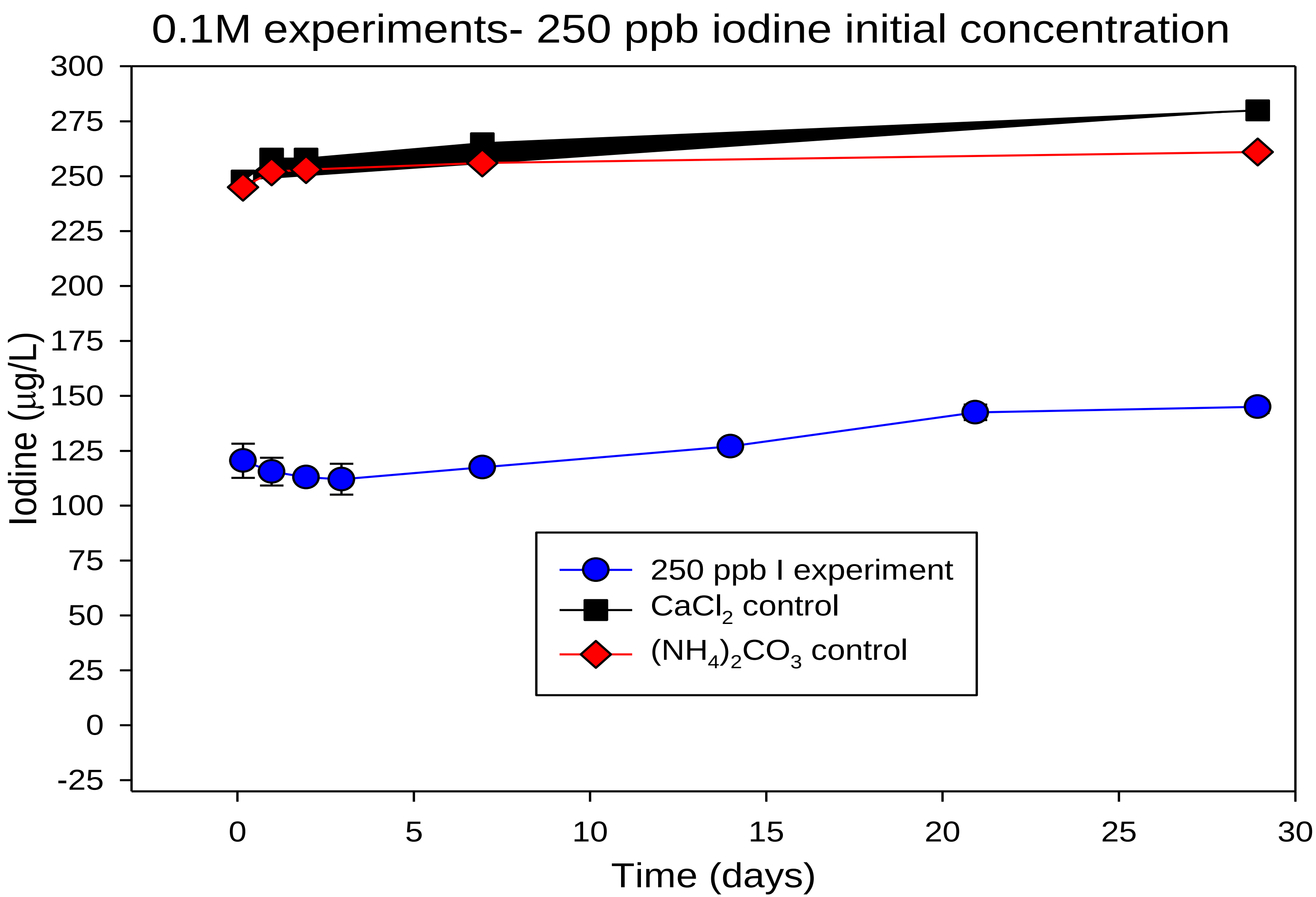

Iodine Geochemistry – Precipitation (Cont.)

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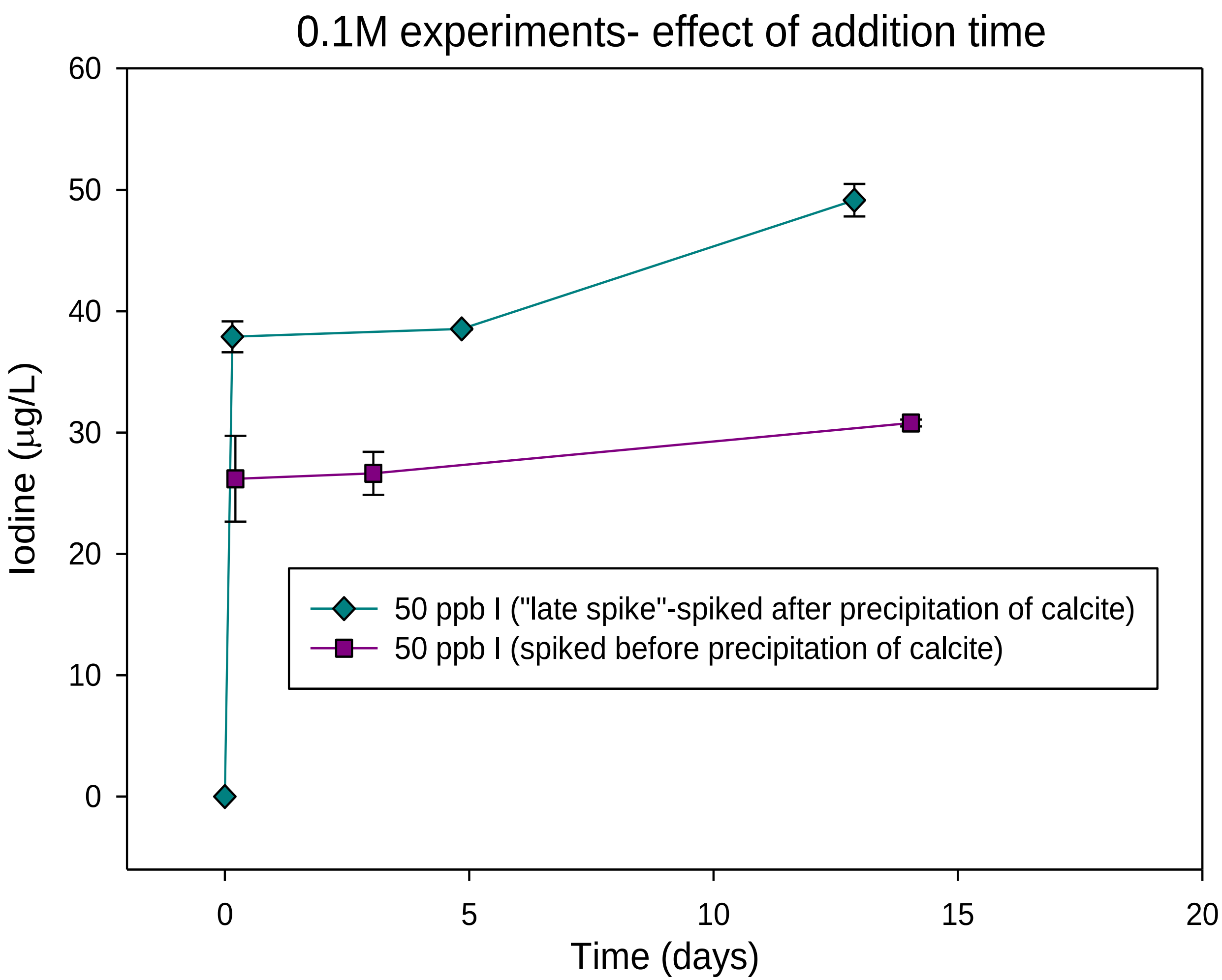
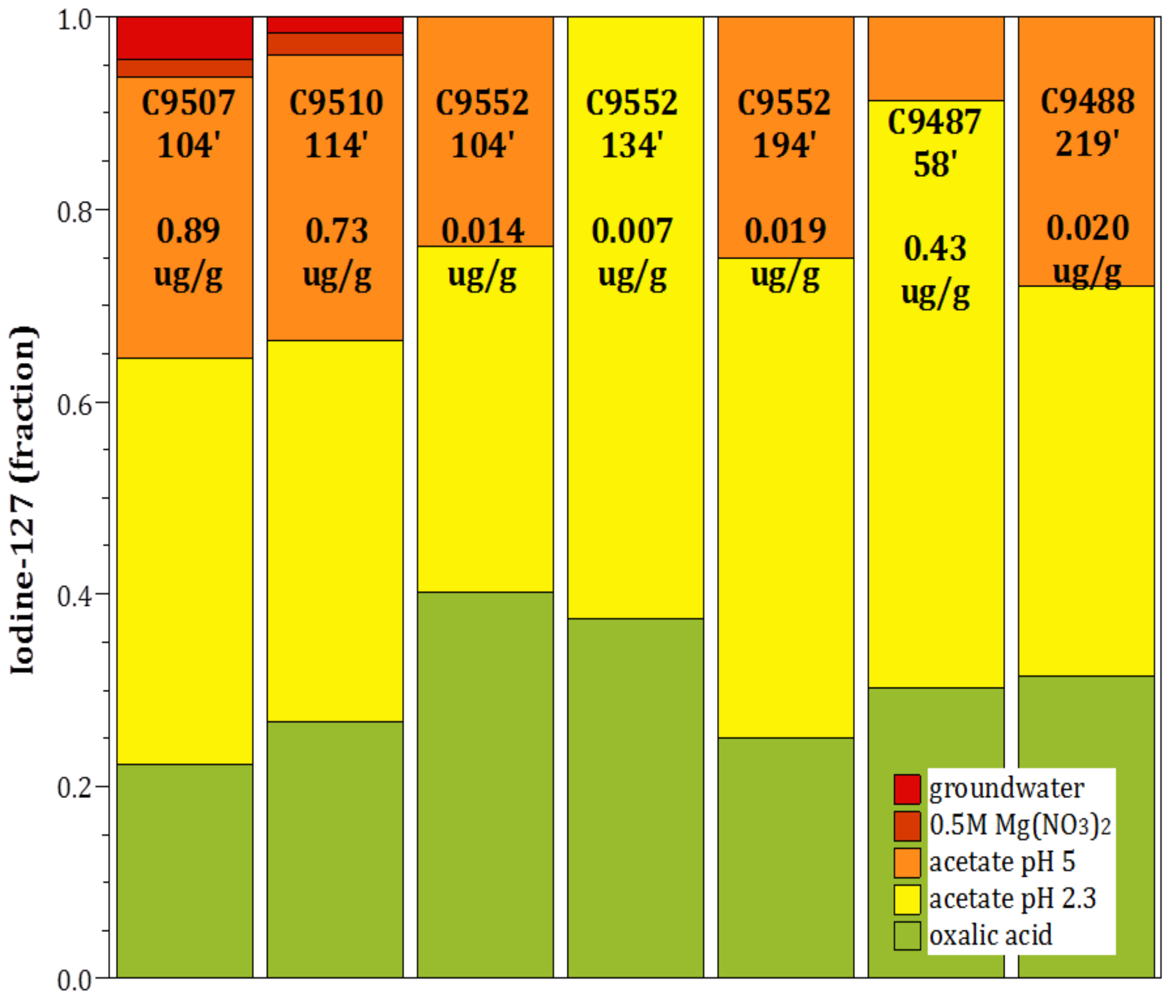
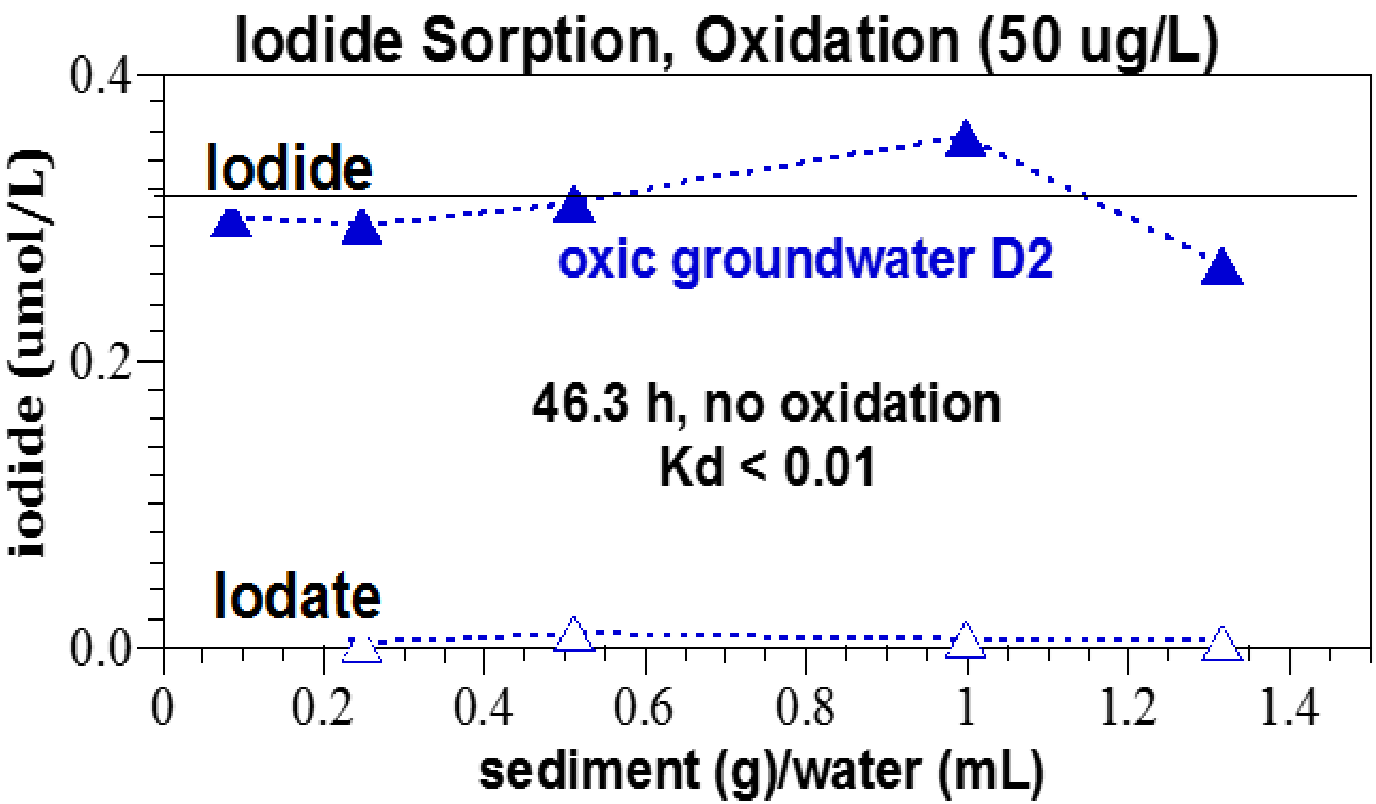


Iodine Geochemistry – Precipitation in Vadose Zone Sediments



Iodine Geochemistry - Adsorption



Iodine Geochemistry – Adsorption (Cont.)

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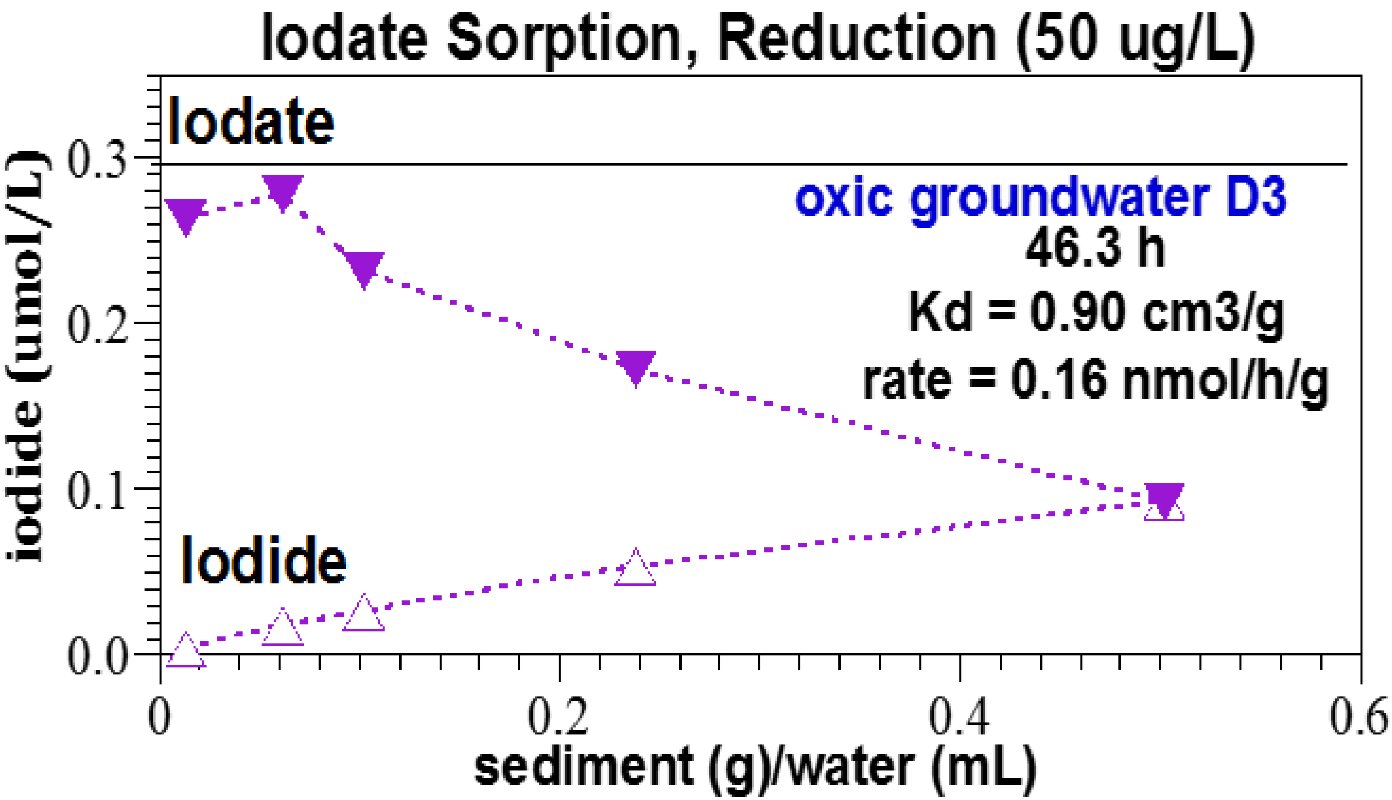
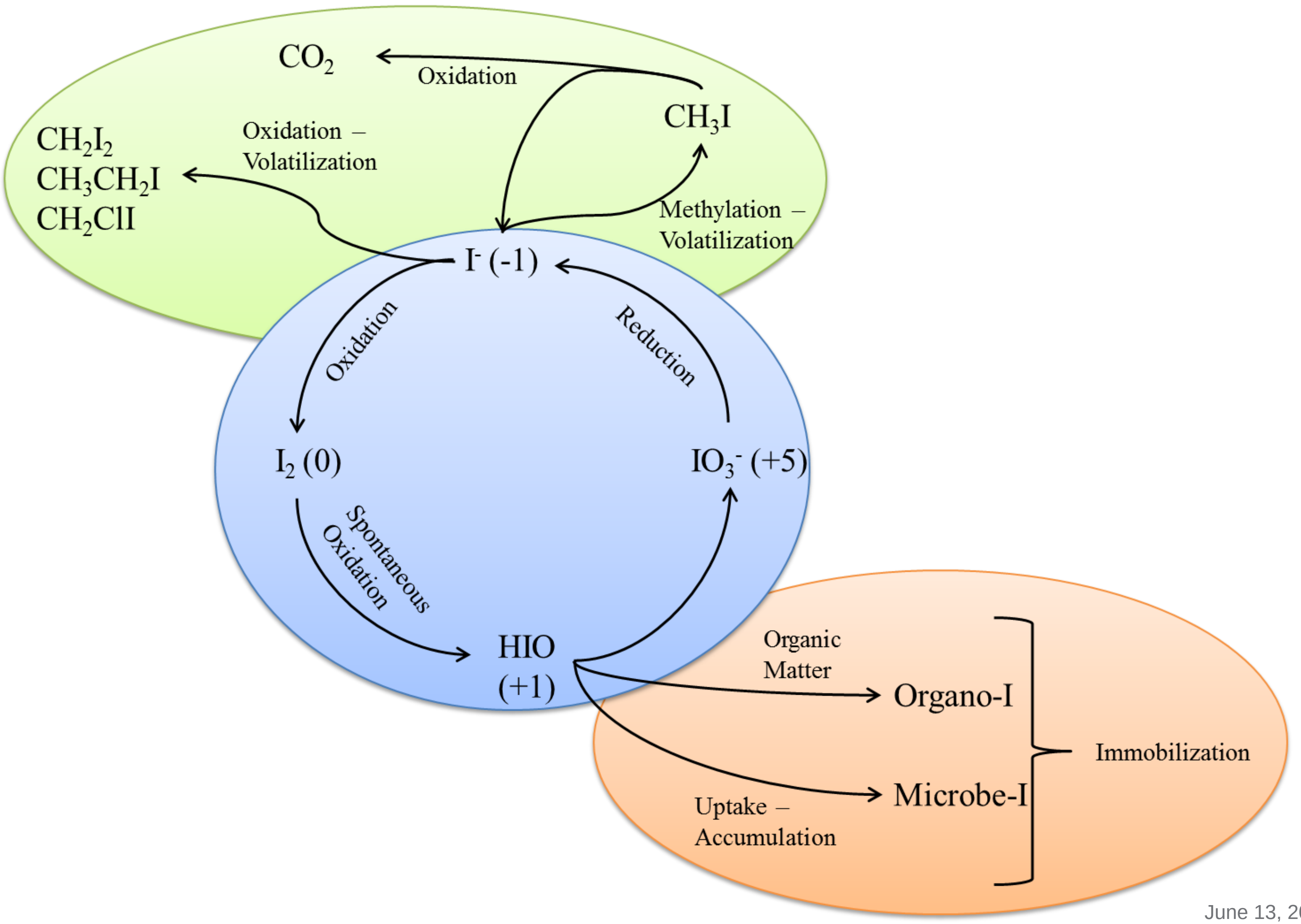


Microbially-Catalyzed Iodine Cycling in the Natural Environment



Installation of Traps in ^{129}I impacted groundwater monitoring wells

^{129}I Concentrations (pCi/L) as a Function of Time

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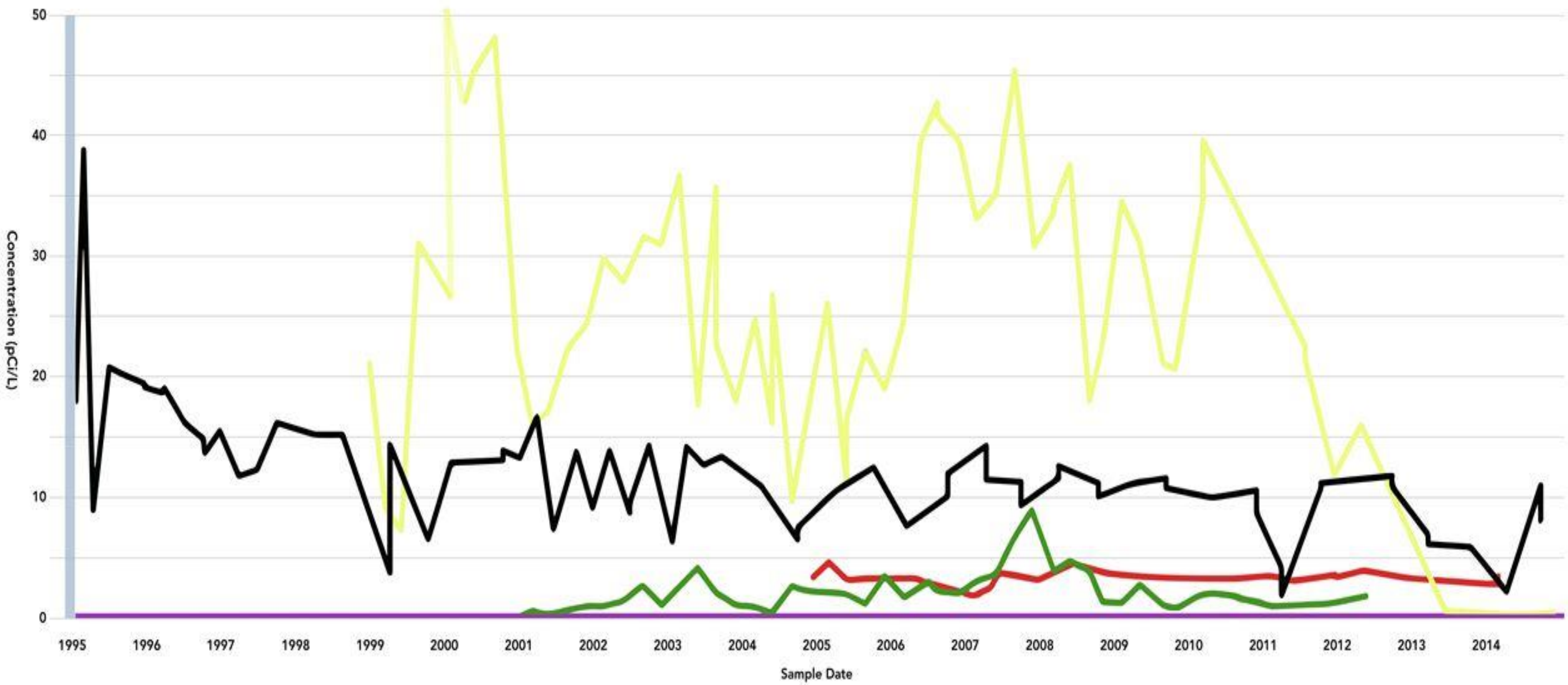
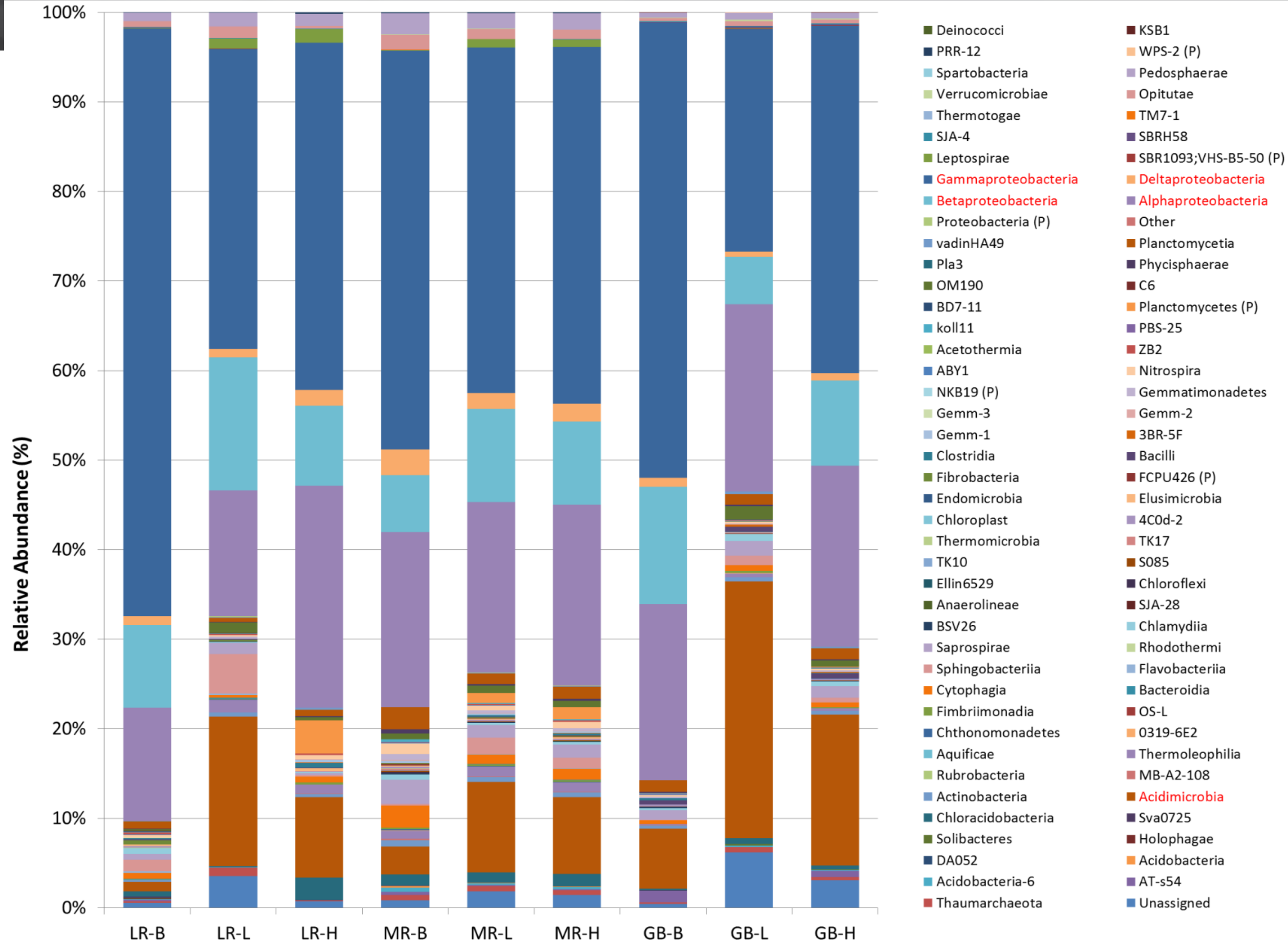


I-129 concentrations in wells: 13 (high I-129; yellow); 15(low I-129; green); 19 (background I-129; purple); 70A (high I-129; black); 70B (low I-129; Red); and 87 (background I-129; not visible) over time

Microbial Diversity from Soils in Traps Deployed in Iodine Contaminated Groundwater - 150 Days



Iodide is Oxidized to Iodine by Multiple Bacterial Species Isolated from Hanford Groundwater

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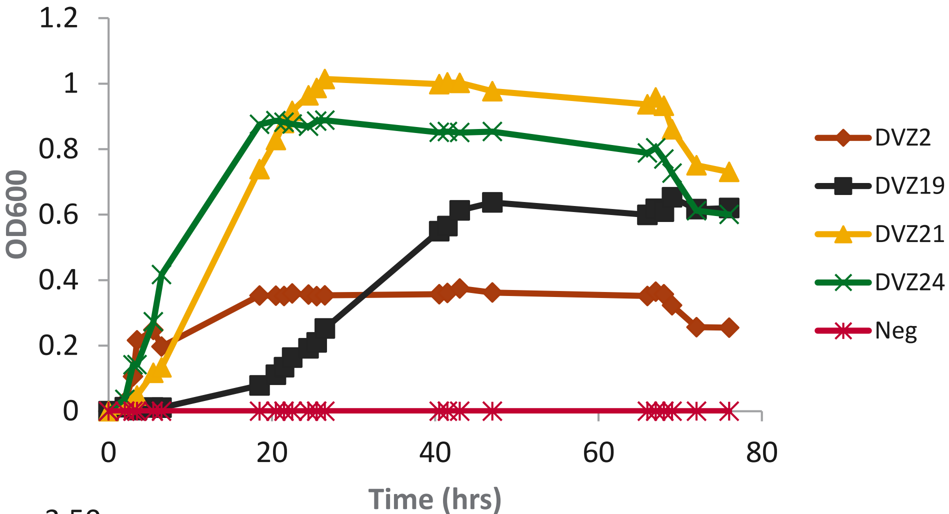
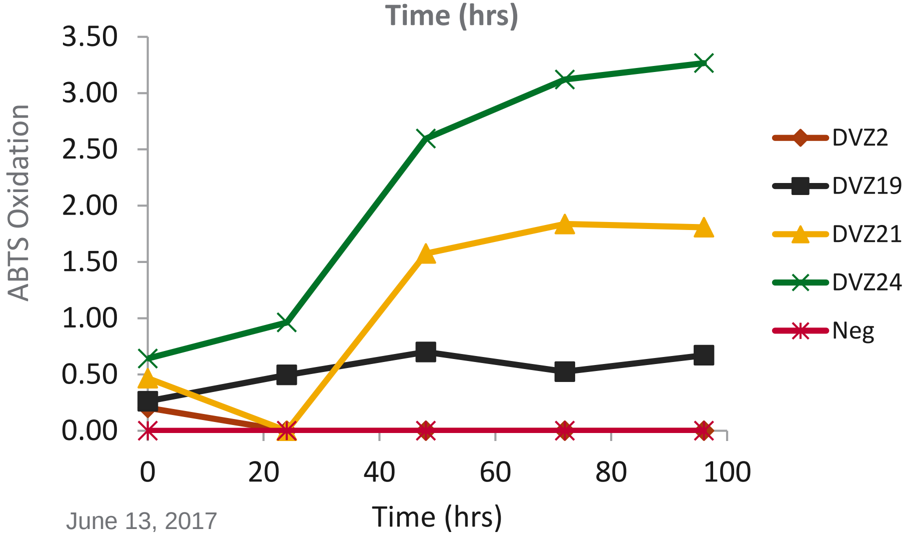
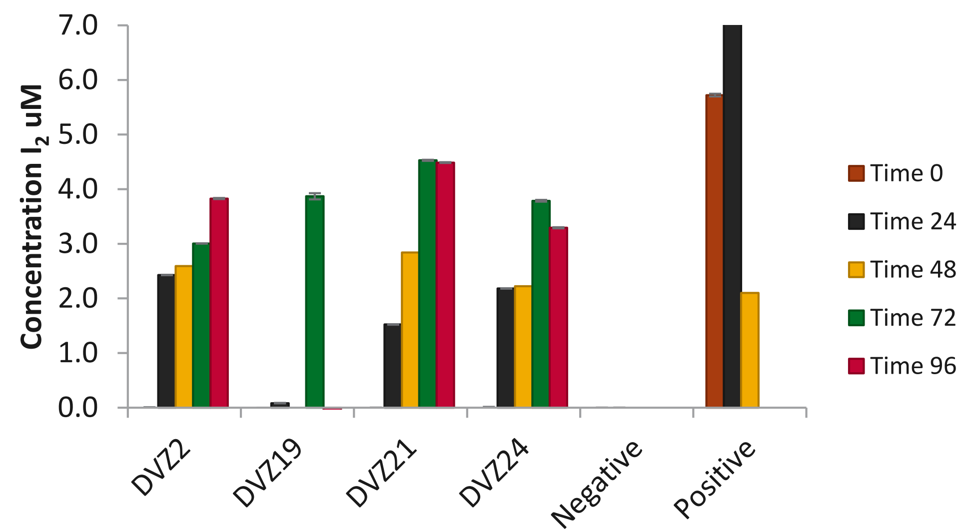
Strain	Affiliation (accession number)
DVZ18	*Pseudomonas* sp. PAMC 26822 (KF011696.1)
DVZ21	*Arthrobacter ilicis* strain ICMP 2608 (NR_104950.1)
DVZ35	*Agrobacterium tumefaciens* strain IAM 12048 (NR_041396.1)
DVZ24	*Pseudomonas mosselii* strain CFML 90-83 (NR_024924.1)
DVZ47	*Shinella kummerowiae* strain CCBAU 25048 (NR_044066.1)
DVZ6	*Pseudomonas mosselii* strain CFML 90-83 (NR_024924.1)
DVZ2	*Bacillus thuringiensis* strain IAM 12077 (NR_043403.1)
DVZ29	*Enterobacter hormaechei* strain 0992-77 (NR_042154.1)
DVZ19	*Microbacterium invictum* strain DC-200 (NR_042708.1)

Bacterial Enzyme Production and Associated Iodide Oxidation

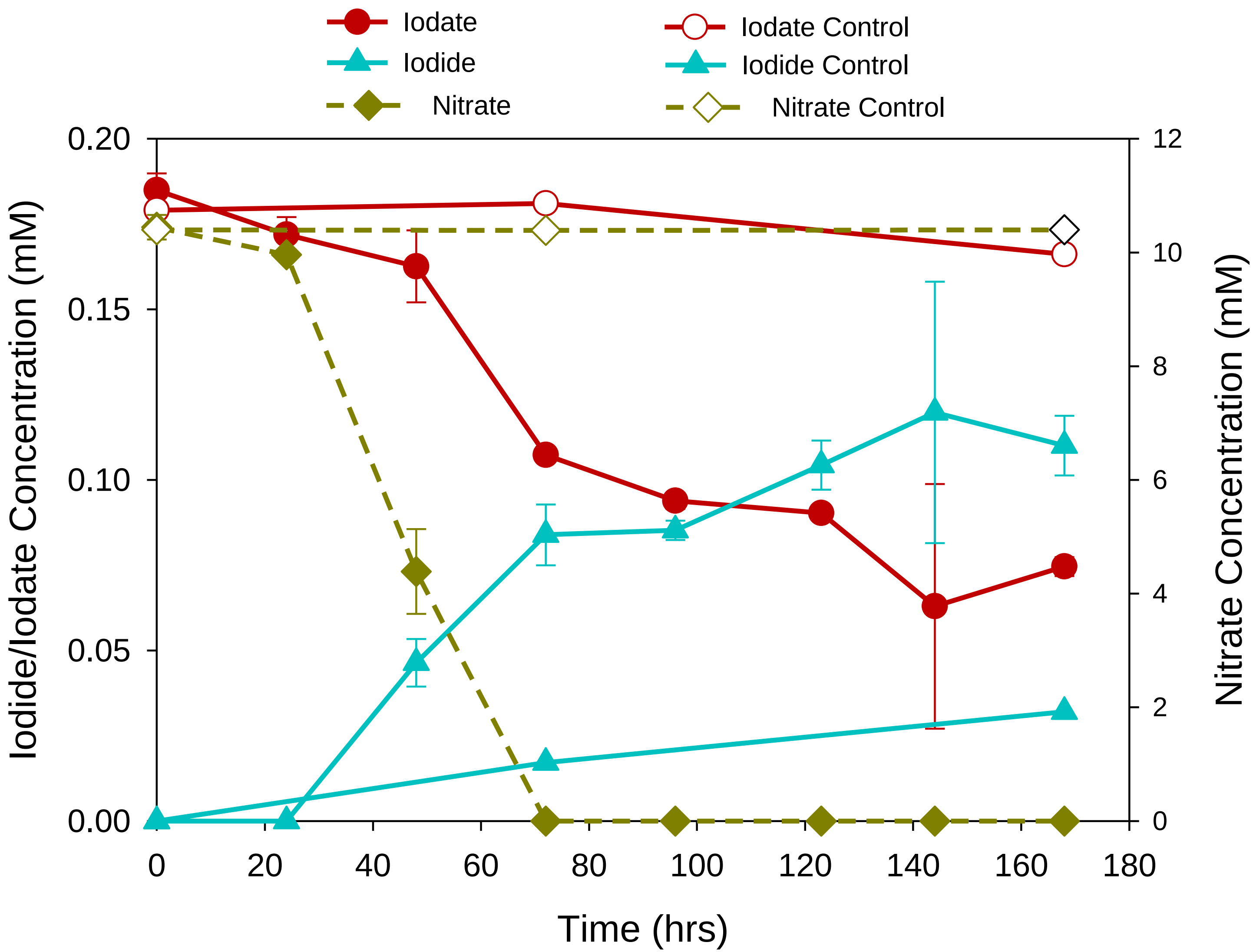
- Bacteria tested demonstrated different growth rates on glucose
- Bacteria produced enzymes known to oxidize iodide
- Iodine production was noted in microcosm studies





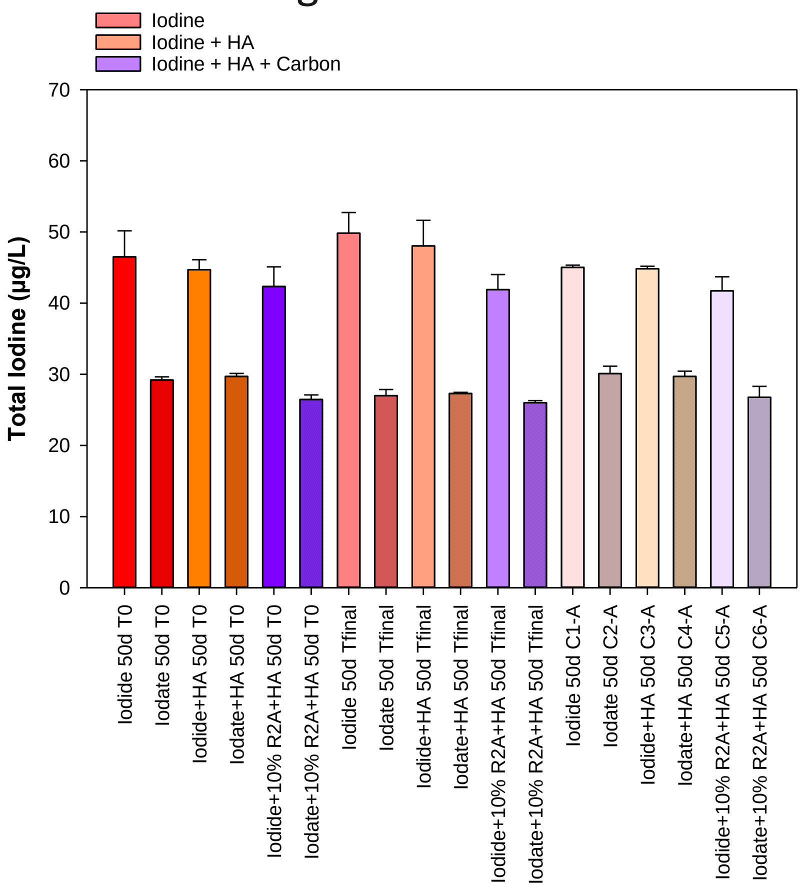


The Dominant Iodine Species (Iodate) is Reduced by Hanford Bacteria

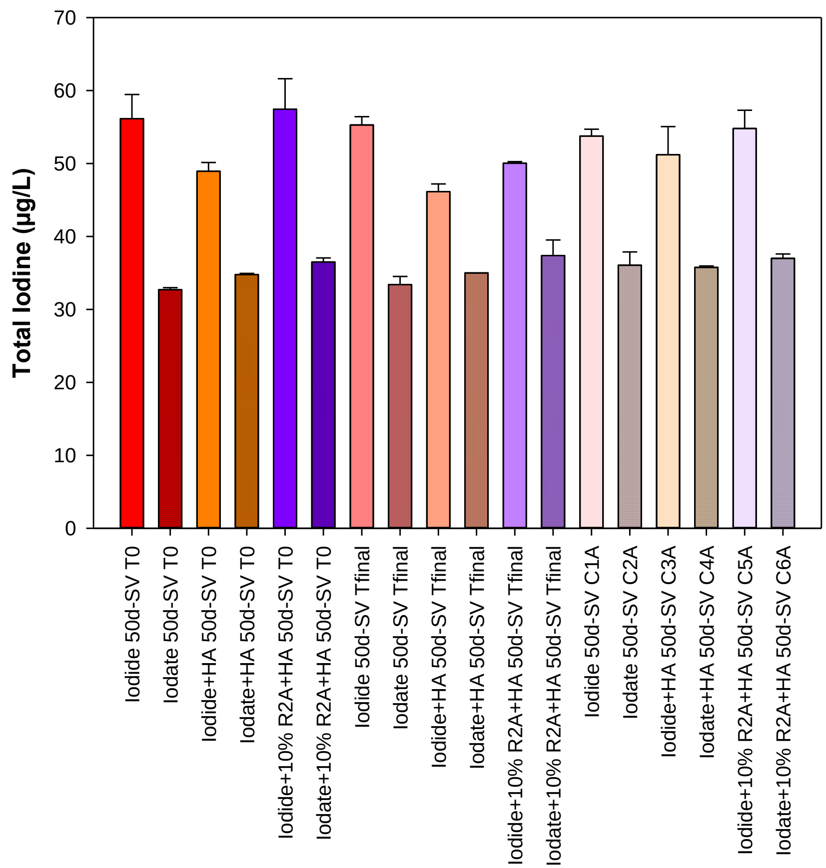


Adsorption of Iodine on NOM

Sigma Humic Acid



Huma K from SRNL



Conclusions

- Numerous controlling processes evident in Hanford groundwater and sediments
  - Microbial community dominated by Pseudomonads
  - Iodate co-precipitates with calcite
  - Iodate is reduced by Hanford sediments
  - Iodine associated with calcite in vadose zone and saturated zone sediments
  - Evidence of biological oxidation and reduction of iodine species by bacteria indigenous to Hanford sediments
  - NOM sorbs iodate to greater extent than iodide
    - Subtle differences between humic acid type
    - Microbial activity didn’t appear to affect adsorption
- Iodate dominates iodine speciation in 200-UP-1 Groundwater

Acknowledgments

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Thank You for Your Attention

Questions?

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