Welcome to the CLU-IN Internet Seminar

Contaminated Sediments: New Tools and Approaches for in-situ Remediation - Session IV

Sponsored by: National Institute of Environmental Health Sciences, Superfund Research Program

February 14, 2011, 2:00 PM - 4:00 PM, EST (19:00-21:00 GMT)

Instructors:

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Moderator:

Steve Mangion, U.S. EPA Region 1 (mangion.steve@epa.gov)

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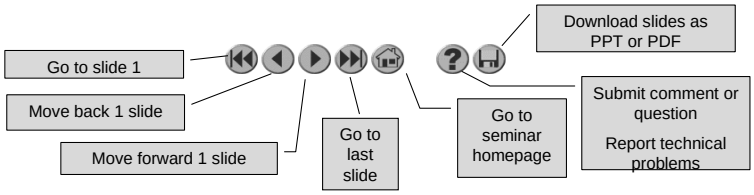

- This event is being recorded
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Integrating Microbial Biostimulation and Electrolytic Aeration to Degrade POPs

Harold D. May, Kevin R. Sowers
Chanlan Chun and Ray Payne

Medical University of South Carolina
And
University of Maryland

February 14, 2011

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Persistent Organic Pollutant: Polychlorinated Biphenyls

Environmental Legacy: Estimated 0.6-1.2 billion kg worldwide

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Goal:

Microbial Degradation of PCBs in Aquatic Sediment

Approach:

1.Identify PCB Dechlorinating Bacteria

2.Develop in situ Monitoring Tools

3.Develop Bioaugmentation

4.Test Bioelectrochemical Stimulation

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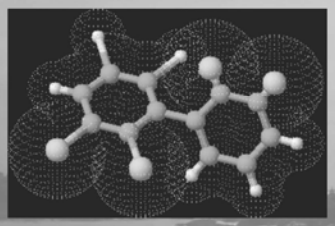
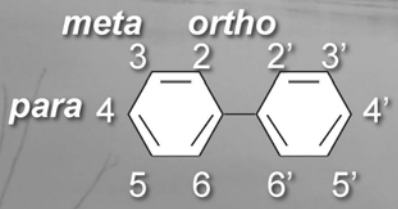
Polychlorinated Biphenyls (PCBs)





- 209 congeners
- Very stable
- Bioaccumulate
- Toxicity concern
- Anoxic sediments are sinks

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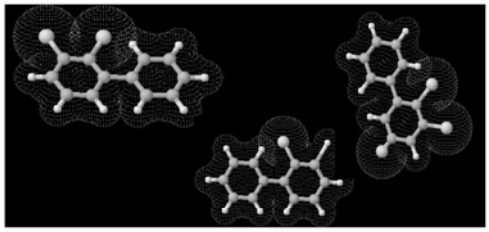
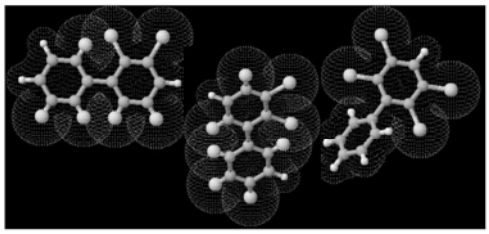
Microbial PCB Degradation



Complementary processes

✓ Aerobic biodegradation
Generally with < 4-6 Cl



✓ Anaerobic dechlorination
generally with ≥ 3 Cl

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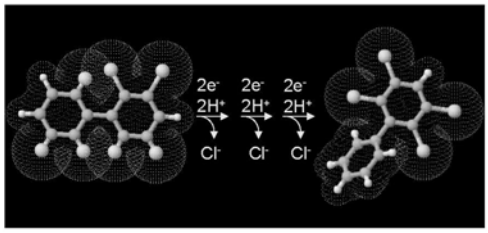
Reductive Dechlorination of PCBs



✓ Performed by anaerobes, e.g. members of the Chlorofexi such as strains DF1, SF1 and *Dehalococcoides* strain CBDB1

✓ Theoretically can produce biphenyl but rarely do. Ordinarily leave mono, di and trichlorobiphenyls behind

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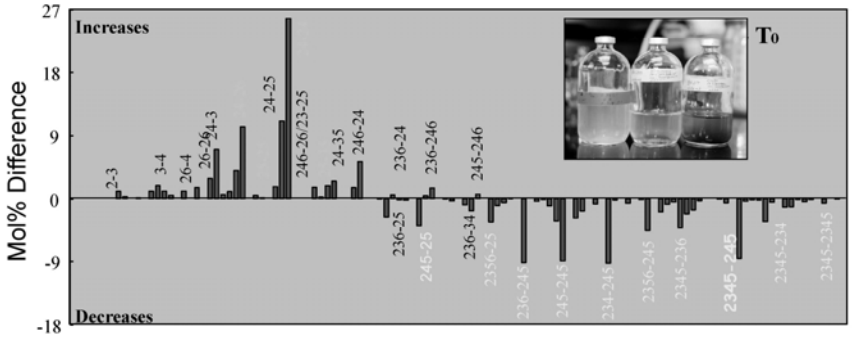
Dechlorination of Aroclor 1260



Wu, et al. *AEM* 1998

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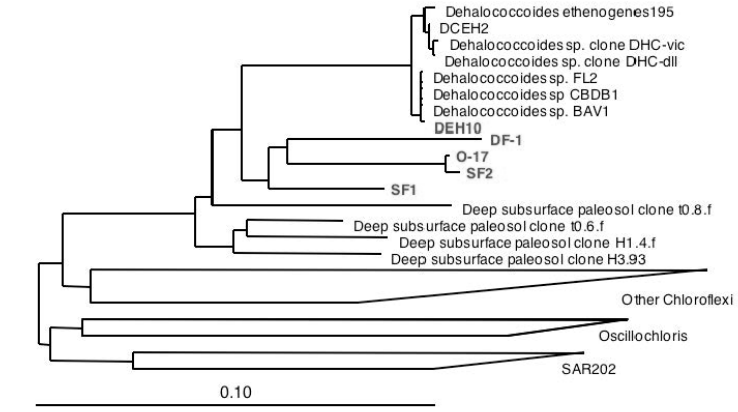
PCB Dechlorinating Bacteria



Based on 16S rRNA gene sequences (1500+ bp)

Cutter et al. *Environ. Microbiol.* 2001, Wu et al., *AEM* 2002, Fagervold et al. *AEM* 2006 &2007, Kjellerup et al. *AEM* 2008

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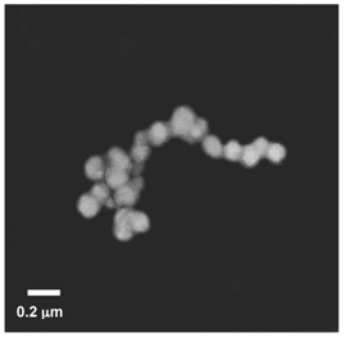
Detecting PCB Dechlorinating Bacteria



- ~~Optical density~~
- ~~Plating~~
- ~~Direct cell counts~~
- ~~FISH~~

May et al, 2008

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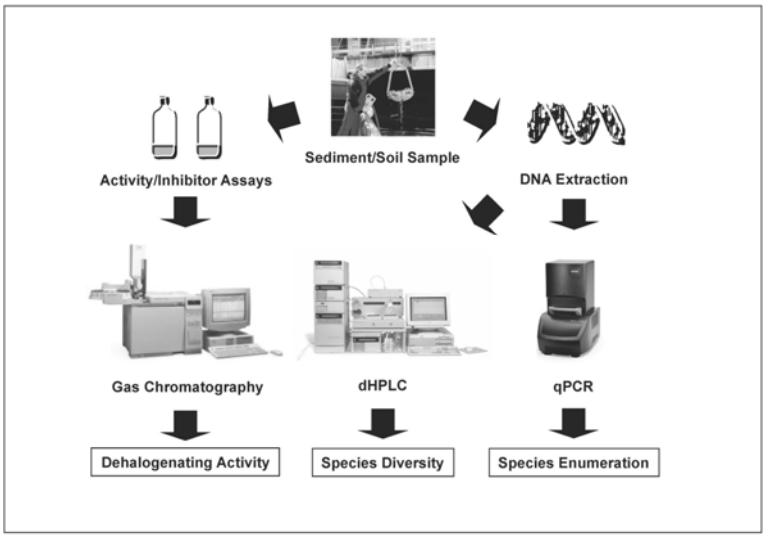
High Throughput Microbial Analysis/Monitoring



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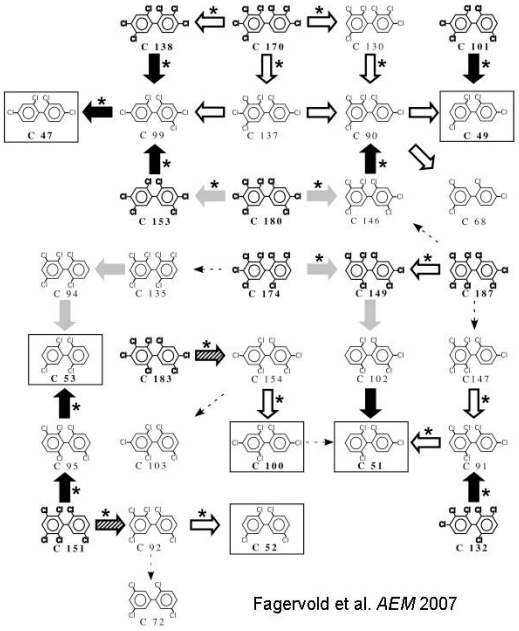
Dechlorination of Congeners Predominant in Aroclor 1260

Only 3 Phylotypes Required

- DEH10
- SF1
- DEH10 & SF1
- O-17



Fagervold et al. *AEM* 2007

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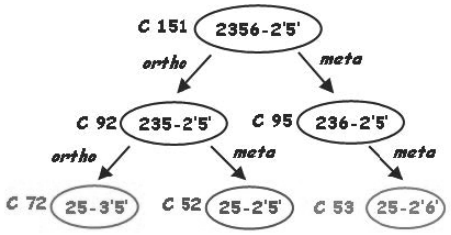
Bioaugmentation with PCB Dechlorinators



o-17,DF1,SF1,DH10	14	46	40
o-17,DF1	54	21	24
SF1,DH10			100
No augmentation	50		50

Fagervold, submitted

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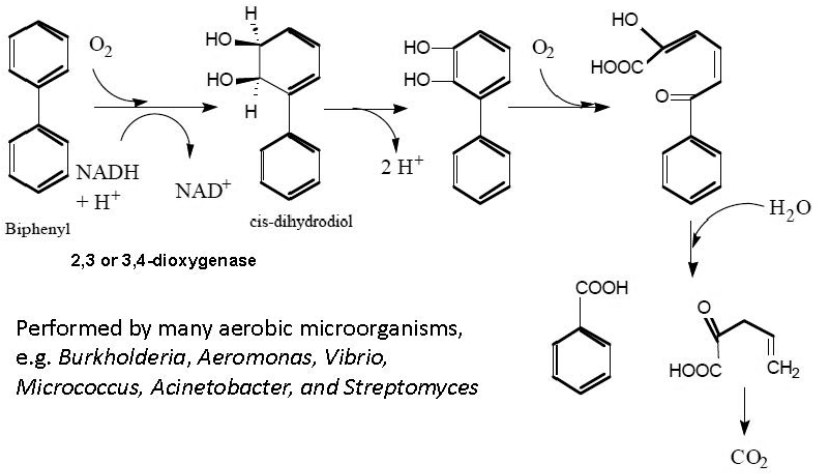
Aerobic Biodegradation of Biphenyl



2,3 or 3,4-dioxygenase

Performed by many aerobic microorganisms, e.g. *Burkholderia*, *Aeromonas*, *Vibrio*, *Micrococcus*, *Acinetobacter*, and *Streptomyces*

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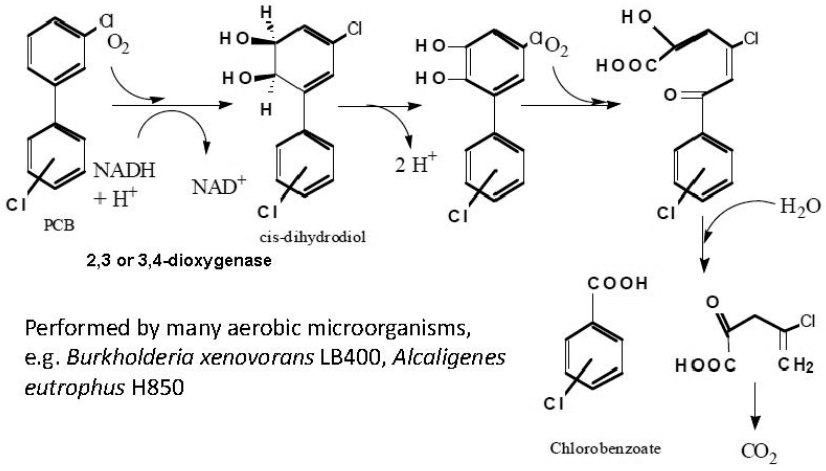
Aerobic Biodegradation of PCBs



Performed by many aerobic microorganisms, e.g. *Burkholderia xenovorans* LB400, *Alcaligenes eutrophus* H850

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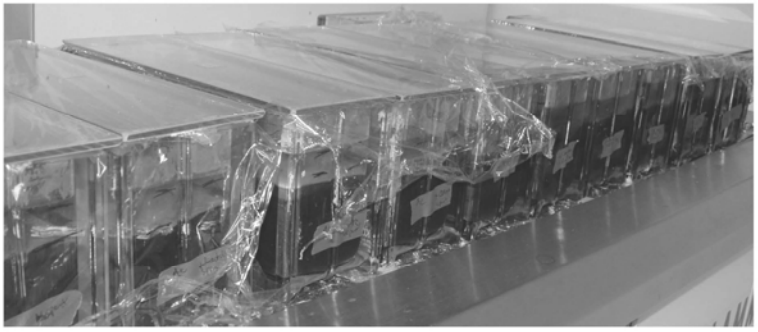
Bioaugmentation of weathered Aroclors

Baltimore Harbor Sediment: 5-10 ppm weathered PCBs



- 1 liter sediment incubated at 20°C in dark
- indigenous water with no additional nutrients
- PCB dechlorinating anaerobe DF1
  and/or PCB degrading aerobe LB400

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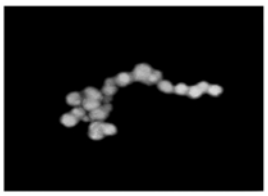
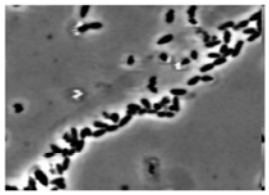
Bioaugmentation of weathered Aroclors



***Dehalohalobium chlorocoercia* DF-1**



***Burkholderia xenovorans* LB400**

- each mesocosm was inoculated with 5 x 10^5 cells
- inoculation by direct injection or on solid substrate
- lactate added as carbon source when indicated

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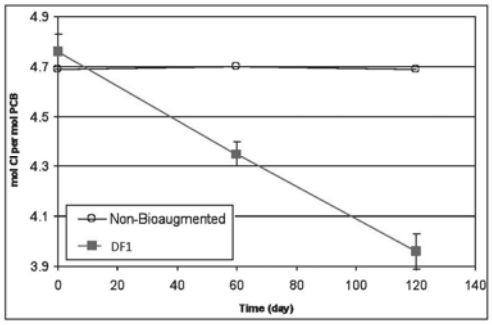
Bioaugmentation of Baltimore Harbor Sediment with DF1



DF1 removed about 0.9 chlorines per mol PCB in 120 days.

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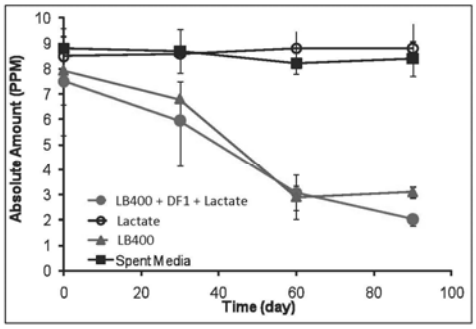
Bioaugmentation with DF-1 + LB400 Results in Degradation of Weathered PCBs



About 75% of total PCBs by mass degraded through oxidative processes and reductive dechlorination in 90 days. Experiment ongoing.

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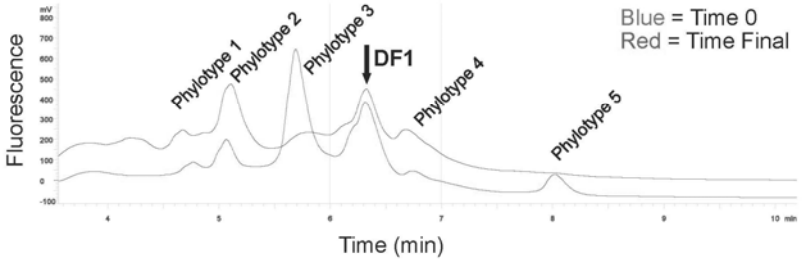
Dechlorinator phylotypes in mesocosm amended with DF1



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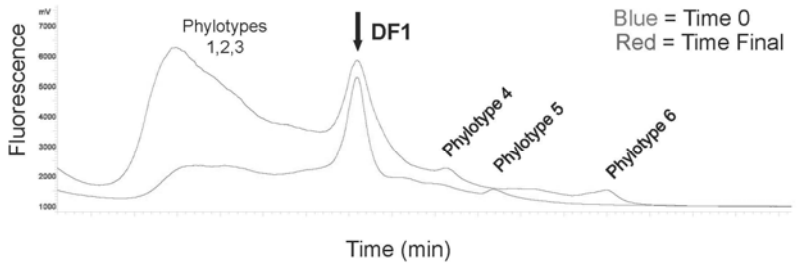
Dechlorinator phylotypes in mesocosm amended with DF1 & LB400



- DF-1 detected by dHPLC at similar levels on days 0 and 90
- Fate of LB400 currently being determined

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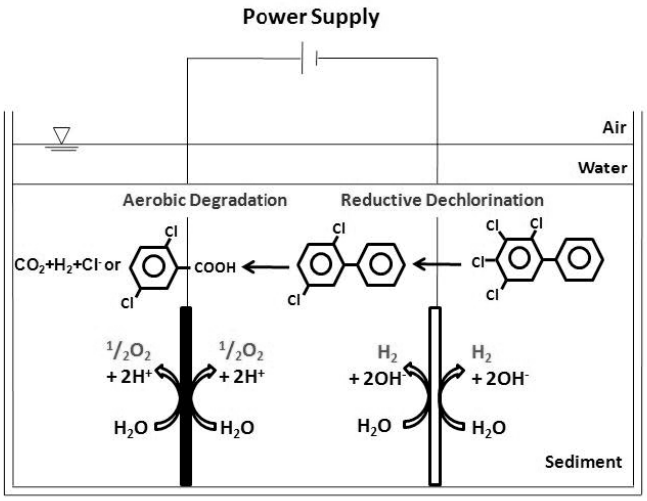
Bioelectrochemical Approach: Hypothesis



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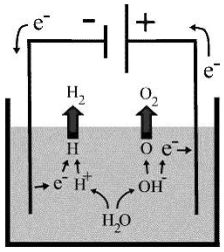
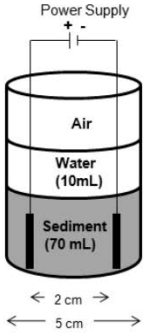
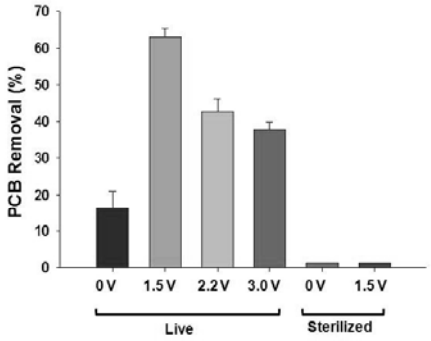
Sediment Bioelectrochemical Reactors (SBRs)

- Microcosm-SBRs
  - Aroclor 1242 impacted sediment (~20 ppm) from Fox River, WI
  - Electrode: Ti sheet (surface area: 12.5 cm^2)
  - Applied voltage: 0, 1.5, 2.2, and 3 V





Sediment Bioelectrochemical Reactors (SBRs)



Up to 65% of total weathered PCBs by mass degraded through oxidative processes in 88 days without bioaugmentation. Increasing chlorobenzoates and benzoate detected with all voltages. Analysis continues.

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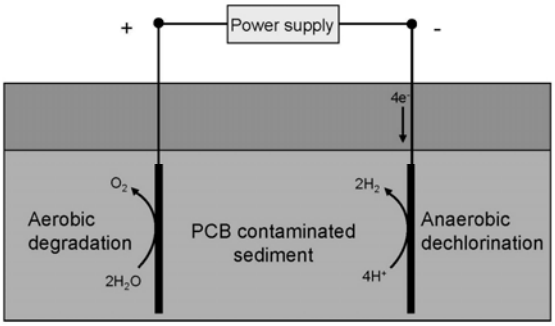
Application of Voltage to Aquatic Sediment



pH problem?

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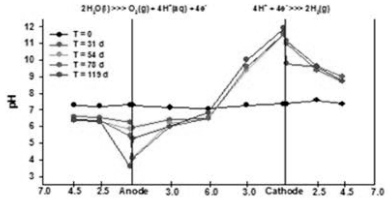
Problem with pH

(Redox, O_2 and H_2 gradients follow suit)

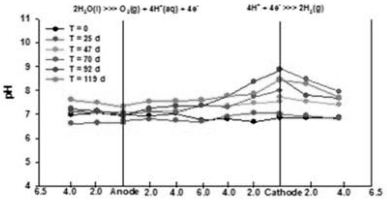
High current density : 0.067 mA/cm^2
(3.8-4.4 V, 15.2-17.6 mW)

Electrode potential vs. Ag/AgCl
Cathode: -1.6 V (H_2 generation)
Anode : 2.5 V (O_2 generation)



Low current density : 0.003 mA/cm^2
(2.0-2.5 V, 0.036-0.045 mW)

Electrode potential vs. Ag/AgCl
Cathode: -1.1 V (H_2 generation)
Anode : 1.4 V (no or slow O_2 generation)



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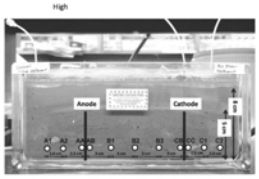
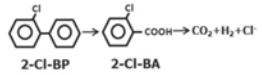
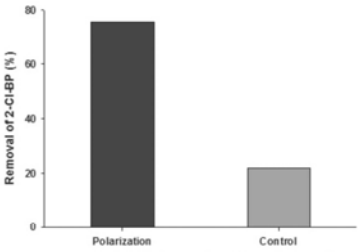
SBR-Polarization

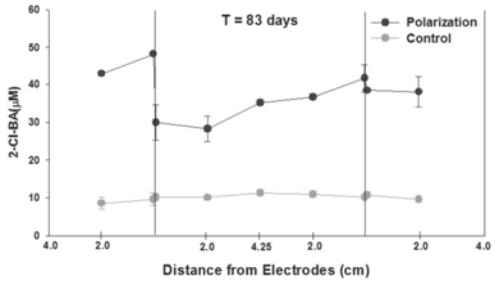
- Mesocosm-SBR
  - PCB-impacted sediment from Fox River, WI
  - Electrodes: Ti sheet (surface area: 55cm^2)
  - Constant Current: 0.014 mA/cm^2, 2.5-3.8 V
  - Model PCB added: 2-Cl-BP





2-Cl-BP 2-Cl-BA





Repeated polarization (cycling of the potential) avoids pH shifts and supports PCB degradation throughout the mesocosm. Weathered PCBs to be tested.

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Conclusions

- Bioaugmentation results in 75% PCB degradation in 90 days
- Application of 1.5V results in 65% PCB degradation
- Anaerobic oxidation of PCBs is hypothesized
- Repeated polarization is successful and avoids pH problems

Future Goals

- Determine full extent of effect of bioaugmentation and electrochemical stimulation
- Determine extent of repeated polarization
- Combine bioaugmentation and electrochemical stimulation
- Test *in situ*

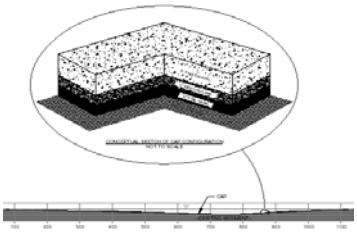
Danny Reible
Environmental and Water Resources
University of Texas

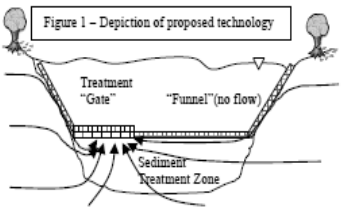
FUNNEL AND GATE APPROACH FOR ACTIVE SEDIMENT CAPS

CO-INVESTIGATORS:
G. LOWRY, K. GREGORY, CARNEGIE-MELLON
J. HUGHES, GA TECH

Sediment In-situ Capping/Treatment

- Reduce risk by:
  - Stabilizing sediments
  - Physically isolating sediment contaminants
  - Reducing contaminant flux to benthos and water column
- Sand effective for strongly solid associated contaminants
- "Active caps" for other situations (w/amendments)
- Funnel and Gate approach to maximize ability to place/replace amendments

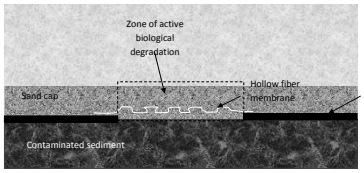


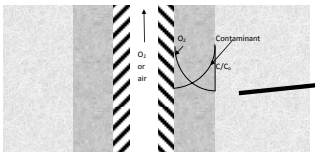


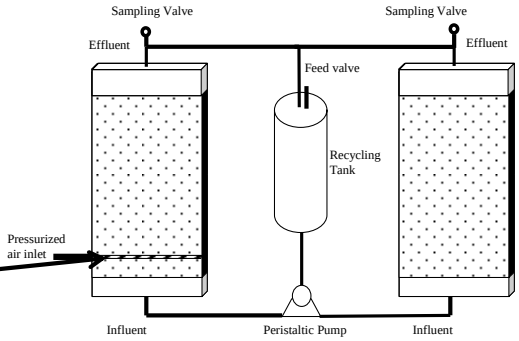
Project Objectives and Scope

- Can a funneling cap effectively contain contaminants and effectively channel interstitial fluids (water or NAPL) to a collection or treatment gate
- Can treatment gates composed of reactive materials effectively manage residual contamination from the in-situ treatment zone
- Scope
  - Preliminary studies with sorbents (e.g. organoclays and activated carbon)
  - Hollow fiber membrane to introduce oxygen in reduced environment
  - Low voltage electrodes to encourage development of appropriate redox conditions

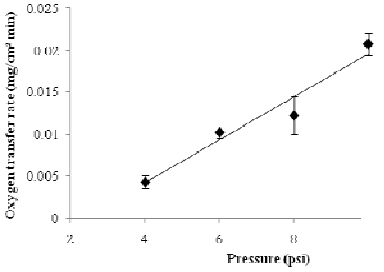
Hollow fiber membrane to introduce oxygen & encourage aerobic degradation







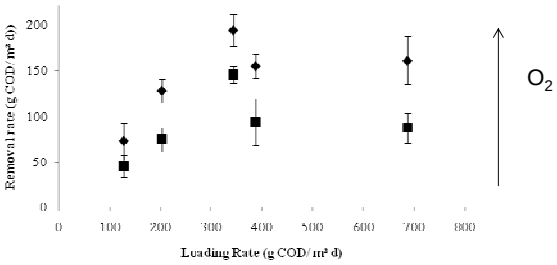
Hollow Fiber Membrane Induced Oxidation



Acetate oxidation

Initially reduced conditions without oxygen introduction via pressurized air

Minimal membrane fouling



O_2

The idea of electro-reactive capping

- Functions:
  - Provide redox control
  - Direct reduction and oxidation on electrode surface
  - Deliver electron donor and acceptor for further degradation
- Advantages:
  - Real time and site-specific control
  - complete mineralization of contaminants through sequential reduction and oxidation

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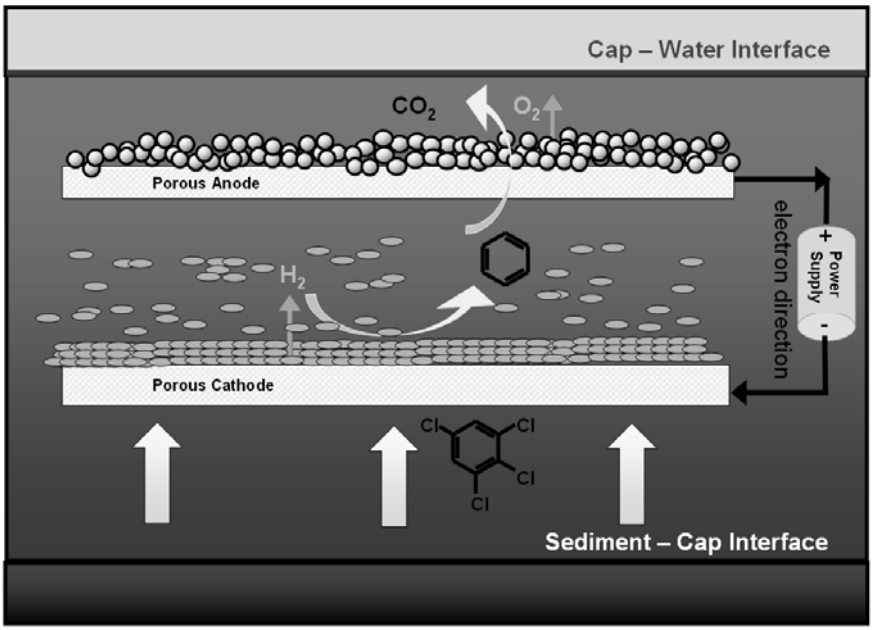
Conceptual model- Electrode cap



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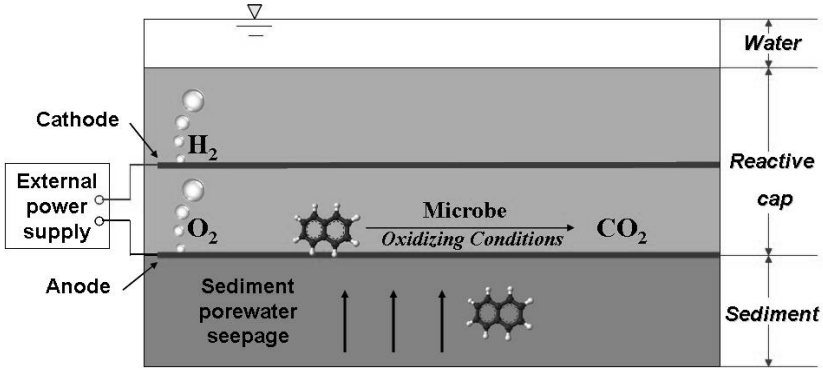
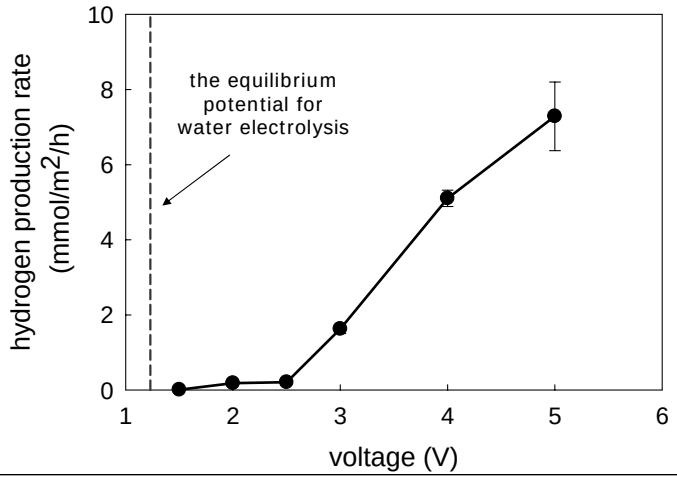
Configuration for PAH oxidation

- Anode at depth to encourage oxidizing conditions where normally strongly reducing conditions exist



Influence of voltage

- Real-time control of electron donor production in the cap by adjusting voltage



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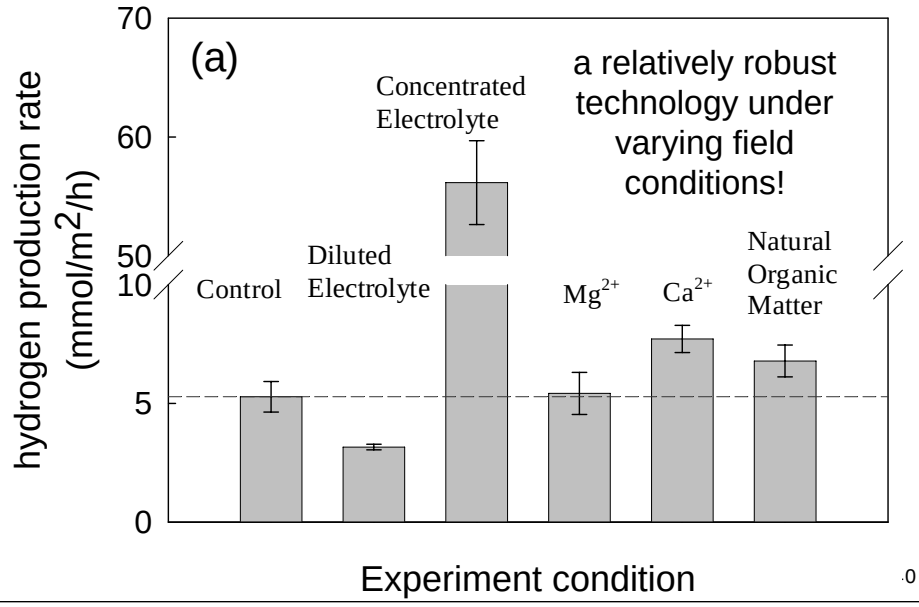
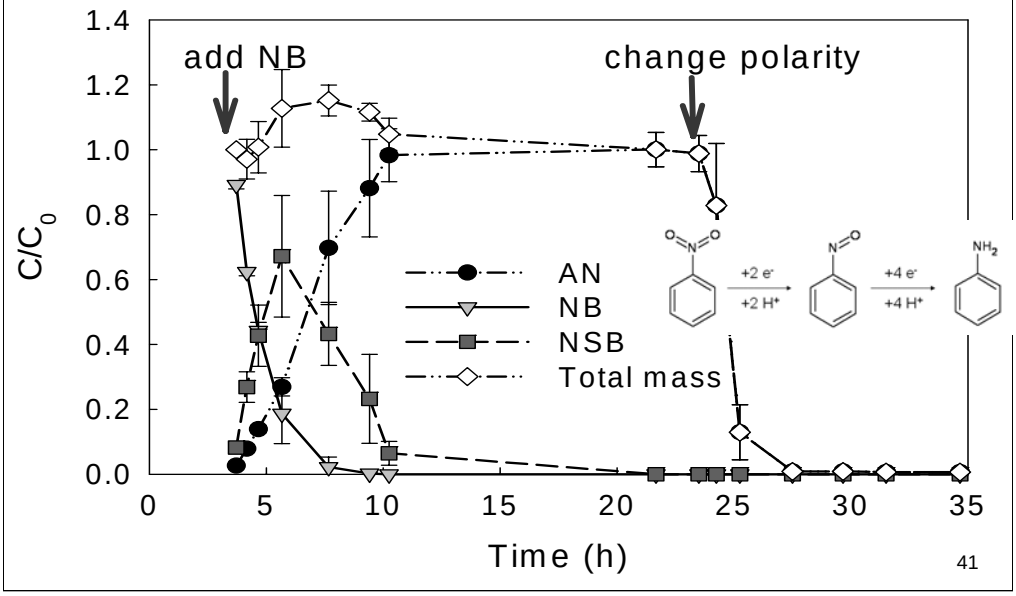
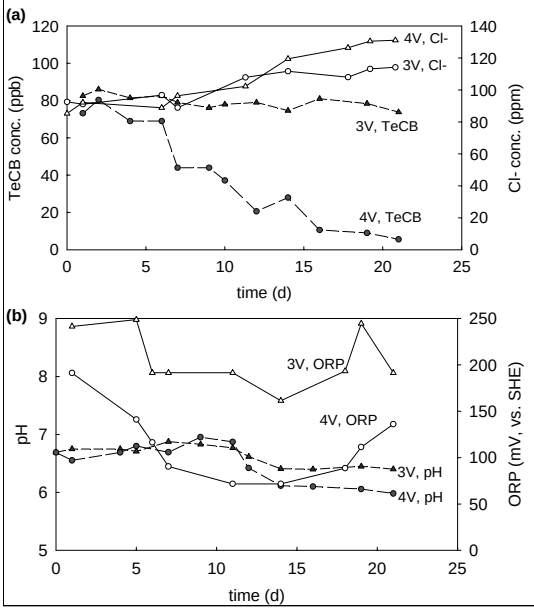
Influence of aqueous chemical species



NB sequential reduction-oxidation



Electrode stimulated degradation of 1,2,3,5- Tetrachlorobenzene



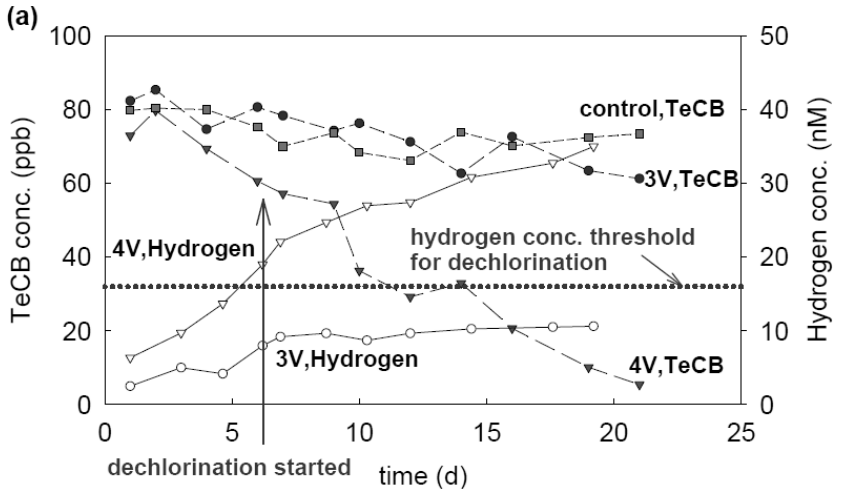
3v insufficient

4v removed ~90% TeCB

ORP: from 200mv to 70mv

pH changes: less 1 unit

Electrode induced dechlorination

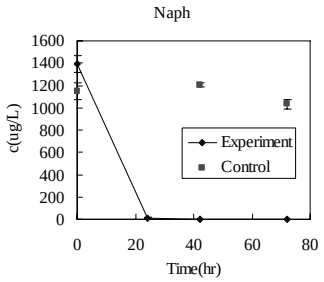


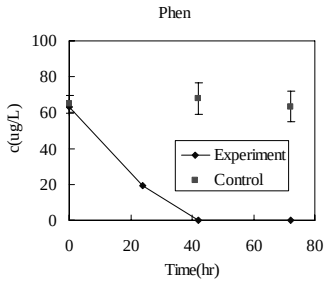
PAH biodegradation in slurry

- PAH degradation under aerobic and nitrate reducing condition
  - To verify PAH biodegradation potential by indigenous microbe in sediment
  - To prove that biodegradation rate is faster in more oxidizing condition (aerobic condition)
- Electrode enhanced degradation of PAH in slurry
  - To examine the feasibility of electrode enhanced biodegradation of PAH in slurry phase

PAH biodegradation under aerobic conditions

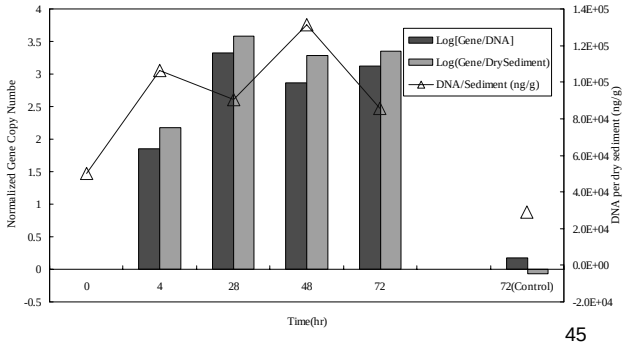
Naphthalene/ Phenanthrene Concentration





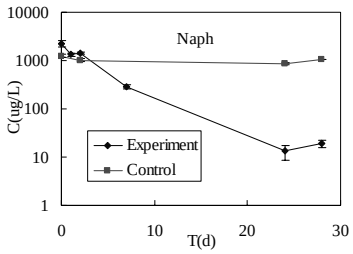
PAH degrading genes

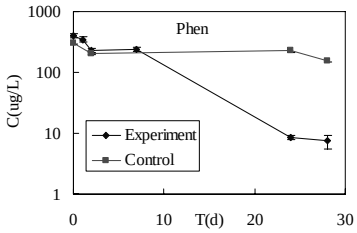
Quantitative polymerase chain reaction (qPCR) using primers that target PAH ring-hydroxylating dioxygenase (PAH-RHD) genes



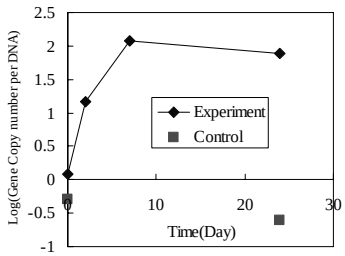
PAH biodegradation under nitrate reducing condition

Naphthalene/ Phenanthrene Concentration





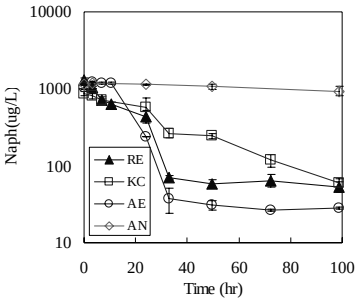
PAH degrading genes

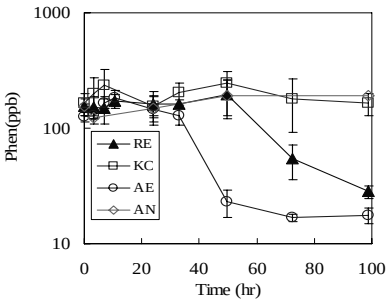


Electrode enhanced biodegradation of PAH in sediment slurry

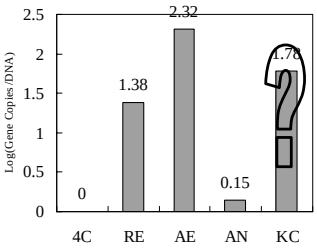
Initially anaerobic slurries

Except fully aerobic slurry





PAH degrading genes



Constant condition slurries

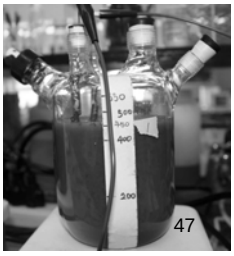
AE: Aerobic condition

AN: Anaerobic condition

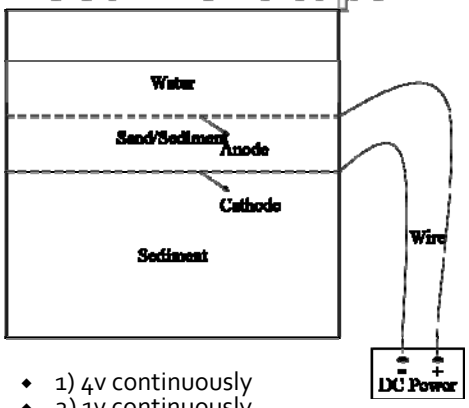
Electrode induced aerobic conditions

RE: Electrode biodegradation

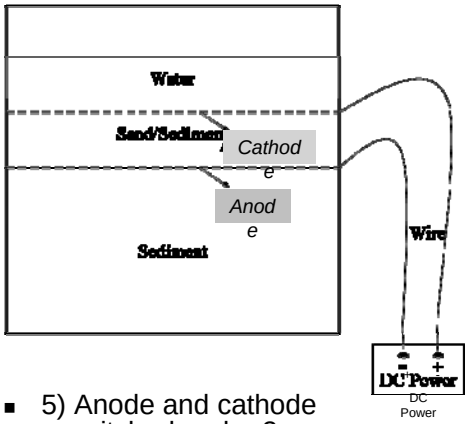
KC: Killed control



Redox control and pH changes in sediment caps with electrodes

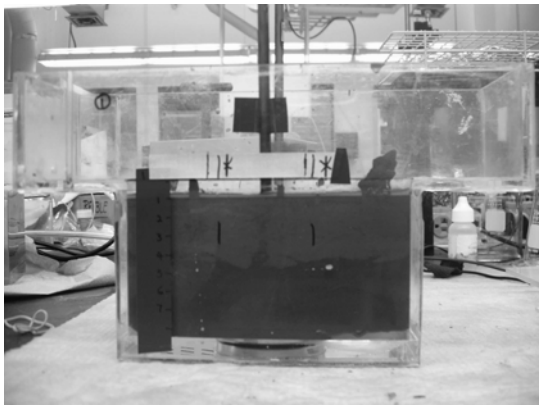


- 1) 4v continuously
- 2) 1v continuously
- 3) 2v continuously
- 4) 4v intermittently (1-3 hours per day)

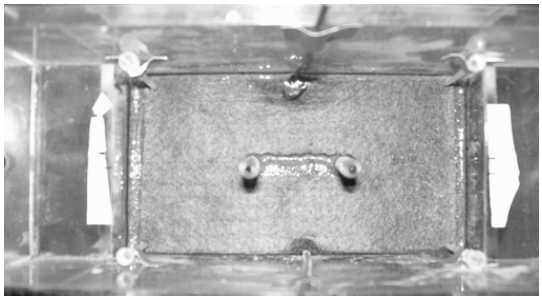


- 5) Anode and cathode switched under 2v

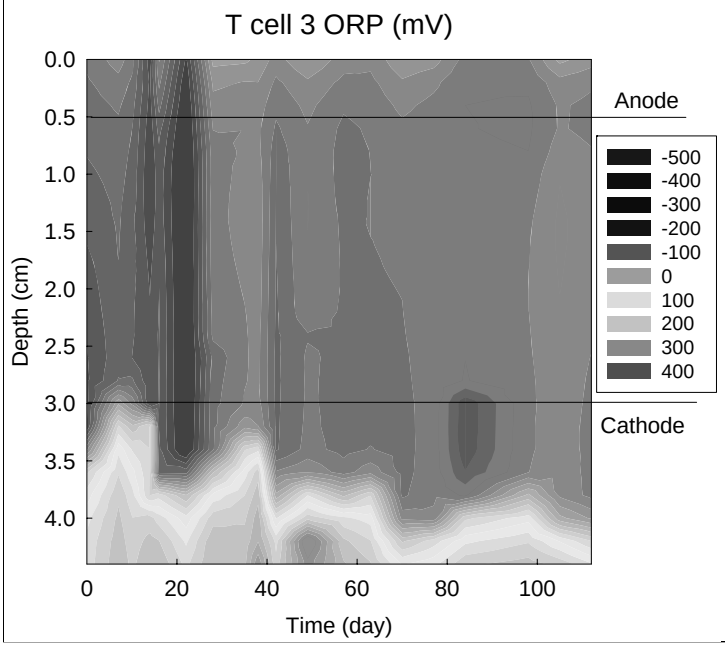
- Side View



- Top View



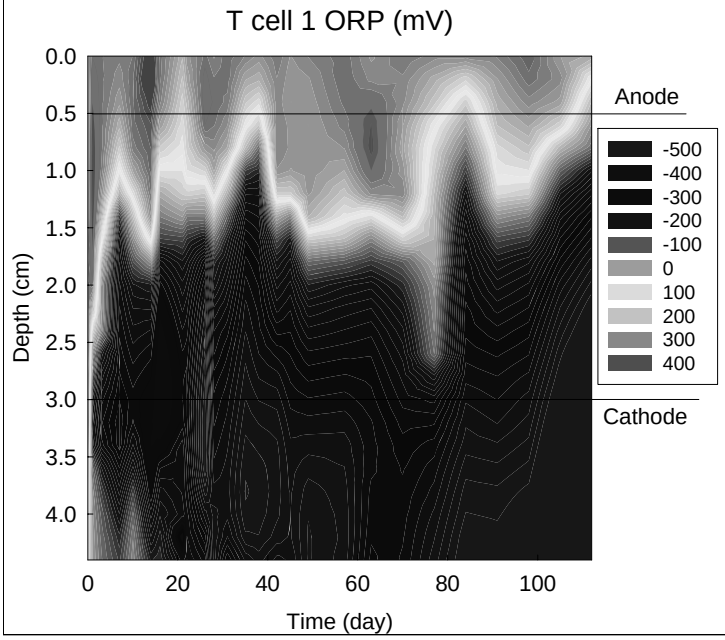
Redox control



Unpowered control

50

Redox control

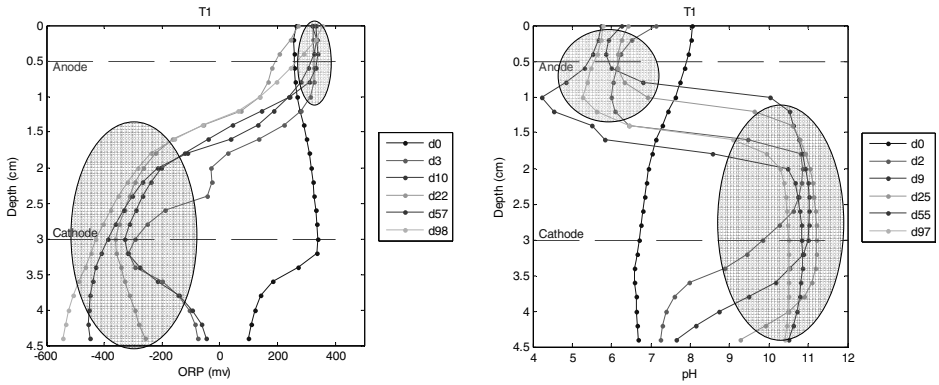


Continuously connected

to 4V DC power supply for~100 days

51

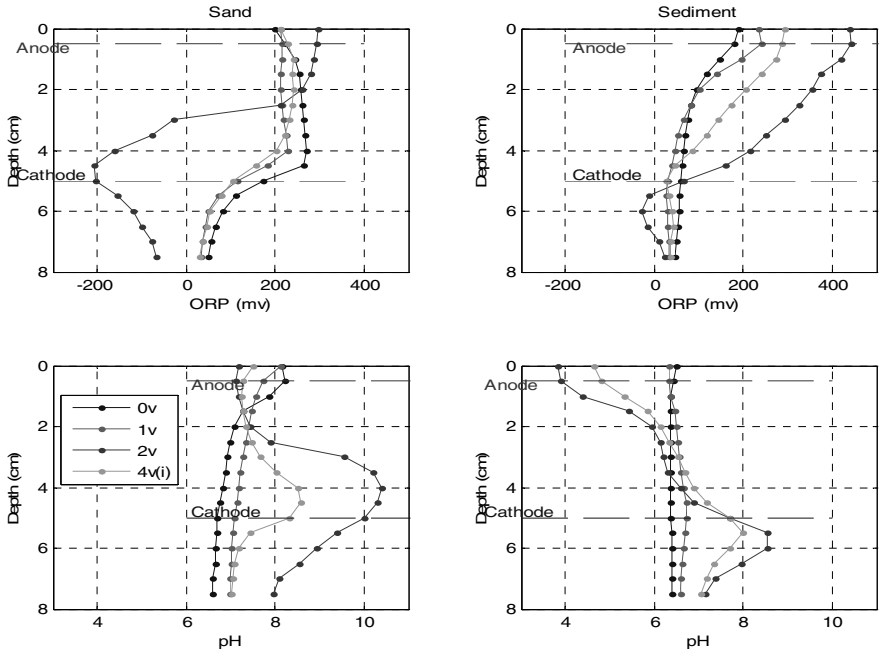
Redox and pH in 4v experiment



Oxidized zone <-> acidic zone

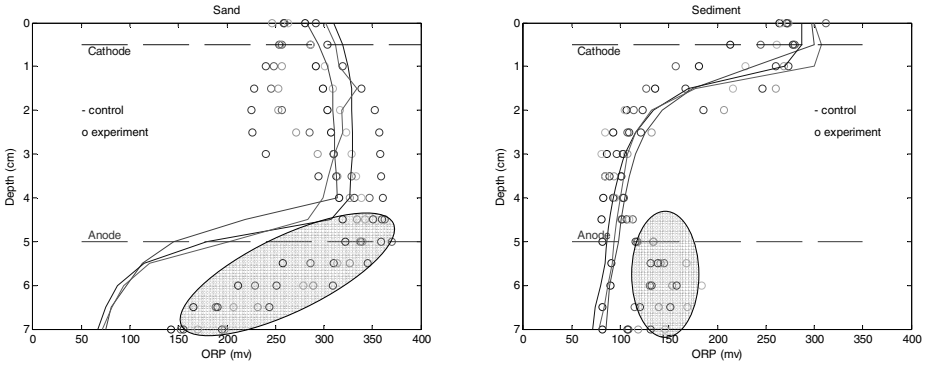
Reduced zone <-> basic zone

Redox and pH in 1v,2v,4v(intermittently) experiment



Redox in *deep oxidation mode*

- Anode at depth (sediment-cap interface)
- PAH degradation mode



Redox control by electrodes

- Proof of concept was successful
- Electrodes could establish oxidizing conditions at the anode
  - Currently studying degradation and increases in PAH degrading genes as per slurry experiments
- pH changes were always associated with redox changes
  - Currently studying use of siderite (iron carbonate) as buffer material to control pH

Intermediate Scale Demonsration

- Large flume demonstrations in cooperation with Bayani Cardenas (UT Geosciences)
- 2 dimensional flow (upwelling, river flow, hyporheic exchange)

Conclusions

- Funnel and gate approach allows significantly more complex and sophisticated capping
  - Regular replacement of finite capacity sorbents
  - Sustainable biodegradation
- Hollow fiber membranes to introduce air/oxygen and encourage aerobic degradation
  - Enhanced degradation shown
- Low power electrodes for encouragement of reduction/oxidation at different layers in sediments
  - Enhanced degradation shown in idealized systems
  - In-sediment studies ongoing
  - Intermediate scale demonstrations planned

Resources & Feedback

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- Please complete the **Feedback Form** to help ensure events like this are offered in the future

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