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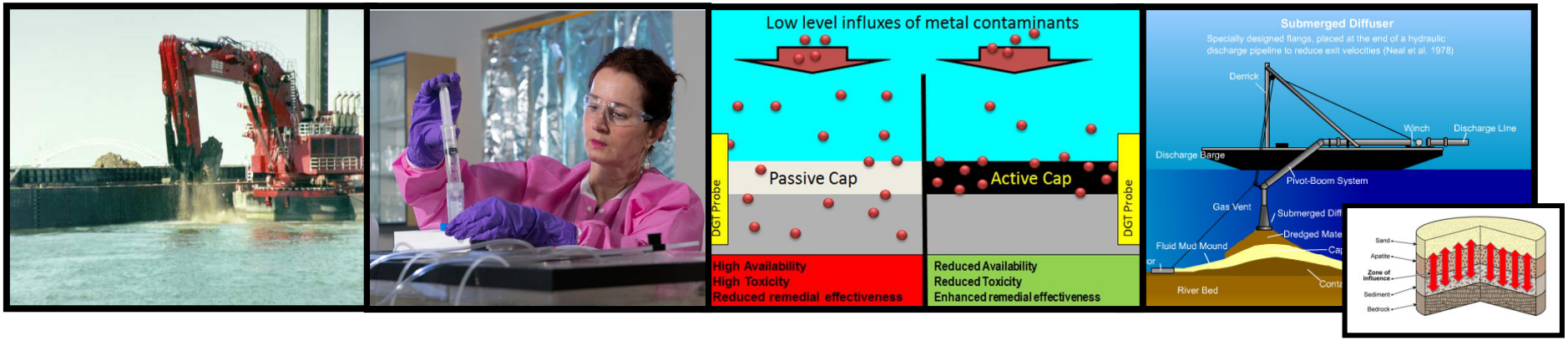
# Environmental Impact of Ongoing Sources of Recontamination on Remediated Aquatic Ecosystems


*Anna Sophia Knox and Michael Paller*

*Savannah River National Laboratory, Aiken, SC, USA*


*February 14, 2019*

*2019 Sediment Conference*

*New Orleans, Louisiana*


SRNL-L3200-2019-00010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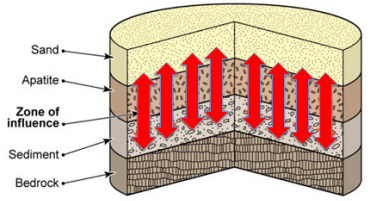
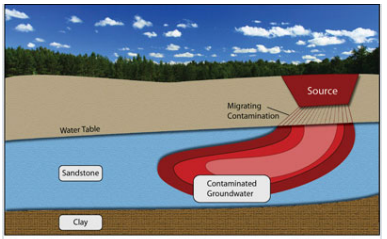
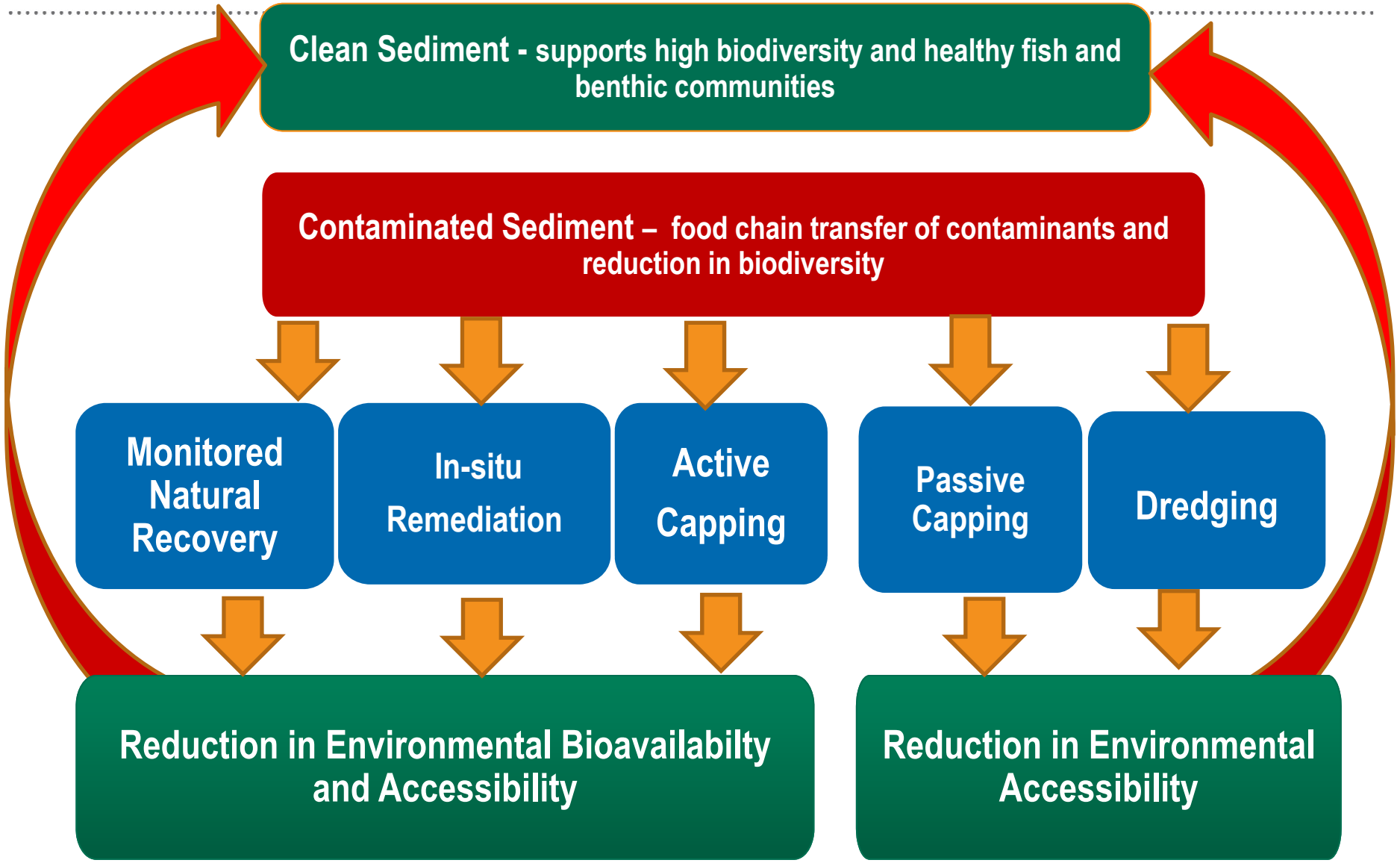
# Problem Statement

- Water is a precious resource supporting life, but only 0.5% of the water is in lakes and rivers and available for human use
- Rapid industrialization and urbanization leads to the contamination of sediments with heavy metals and organic contaminants and creates a pervasive problem worldwide
- Ten percent (1.2 billion $yd^3$) of the sediment in U.S. waters is contaminated (PAHs, PCBs, metals, metalloids, and others)
- Contaminants pose a high risk to the environment and human health because they can harm aquatic organisms and enter aquatic food chains that lead to humans

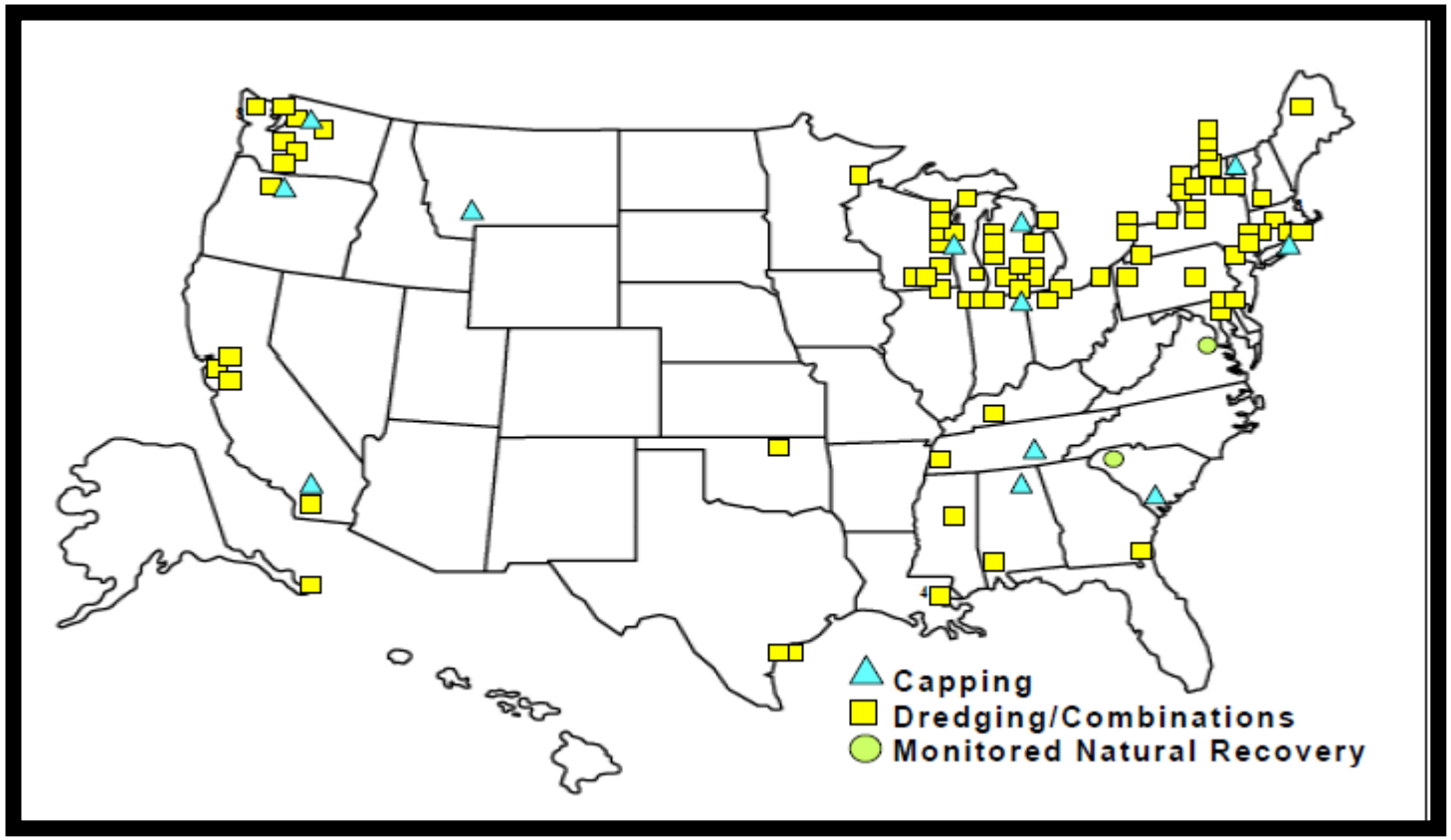




Conceptual model of contaminant plume flowing from a source area and into the water table


Clean Sediment - supports high biodiversity and healthy fish and benthic communities
Contaminated Sediment – food chain transfer of contaminants and reduction in biodiversity
Monitored Natural Recovery
In-situ Remediation
Active Capping
Passive Capping
Dredging
Reduction in Environmental Bioavailabilty and Accessibility
Reduction in Environmental Accessibility

# Distribution of Completed and On-going Projects in the U.S. with Greater than 2000 Cubic Meters of Contaminated Sediments

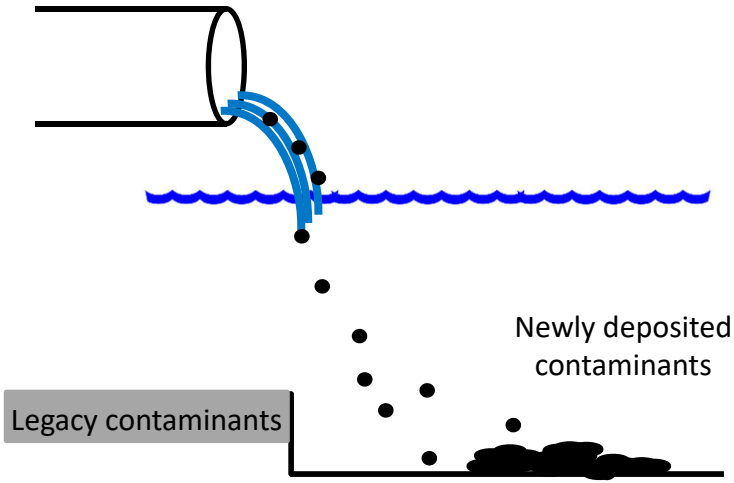


# Recontamination – a Challenge to Remedial Methods for Sediment

**Remediated sediments may be exposed to contamination from uncontrolled point or nonpoint sources resulting in recontamination that reverses recovery.**

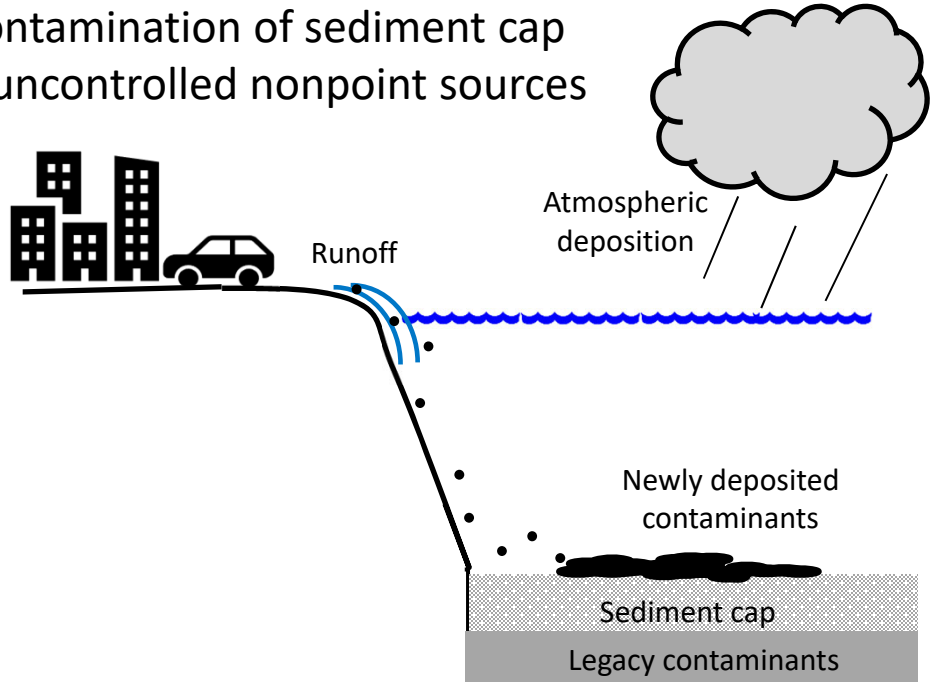
**The effects of recontamination on sediment remediation have not been evaluated**

Contamination of dredged area by uncontrolled point source



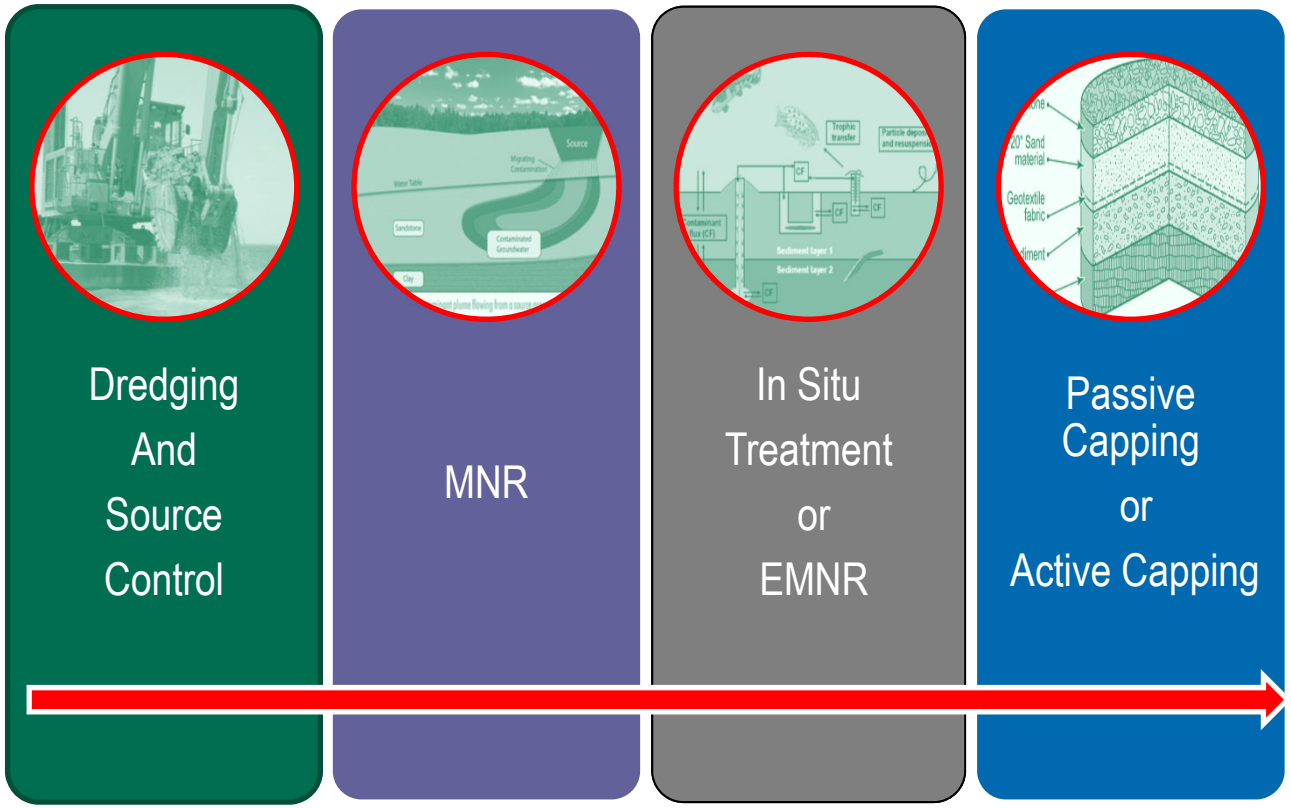

Benefits of dredging to remove legacy contaminants are negated by recontamination!

Contamination of sediment cap by uncontrolled nonpoint sources




Benefits of sediment capping to isolate legacy contaminants are negated by recontamination!

# Logical Progression in Remediation of Contaminated Sediments in Case of Controlled Sources

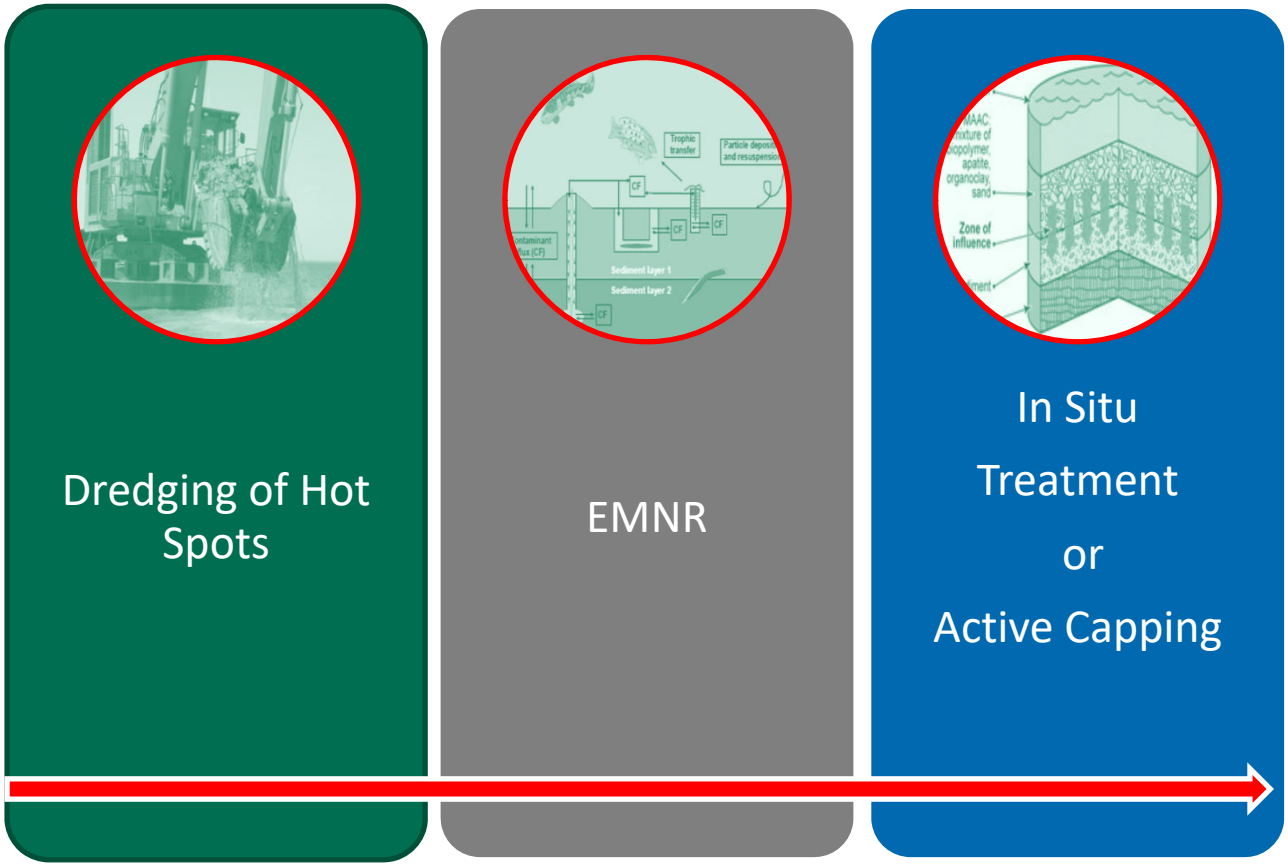



After dredging and source control additional engineering can be added if more than MNR is required

(Bridges et al., 2012. Integrated Environmental Assessment and Management 8: 331-338)

MNR - Monitored Natural Recovery
EMNR – Enhanced Monitored Natural Recovery

# Logical Progression in Remediation of Contaminated Sediments in Case of Ongoing Sources

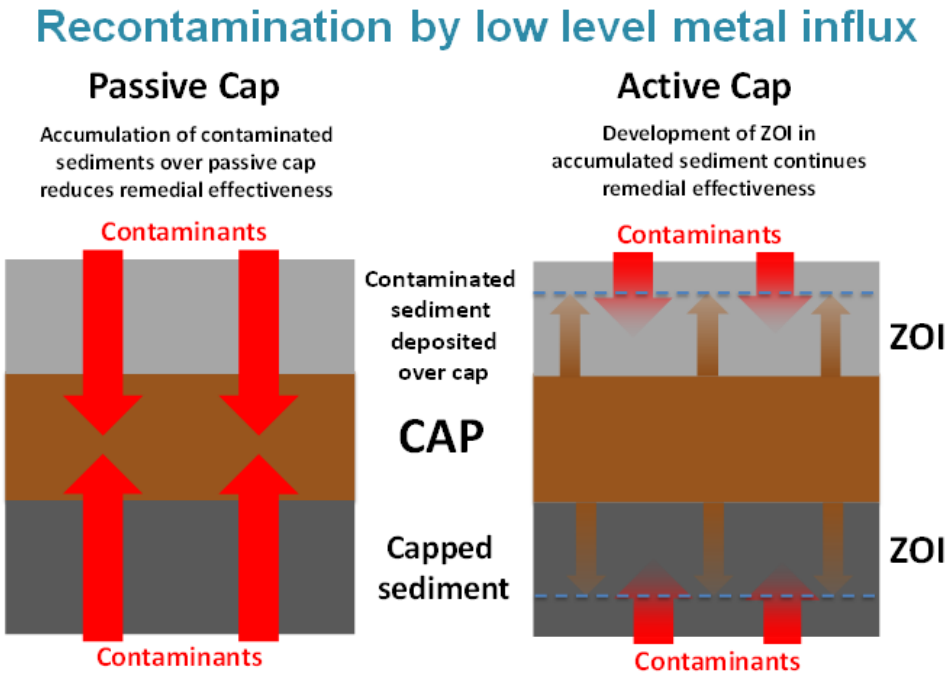



**EMNR – Enhanced Monitored Natural Recovery**

# Technologies that Remediate Existing Contaminants in Sediments and Control/Remediate Ongoing Sources are Needed

## Passive versus Active Capping

**Passive Cap**
**Layer of inert material (e.g., sand) that isolates contaminated sediment from the surrounding environment**



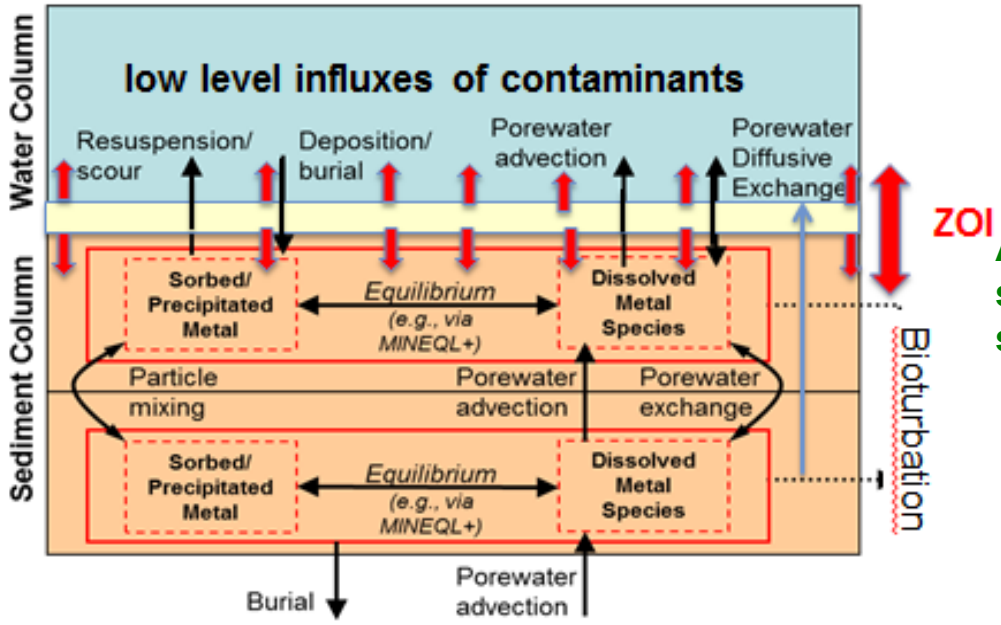

**Active Cap (single amendment)**
**Reactive material that neutralizes or stabilizes contaminants in situ**

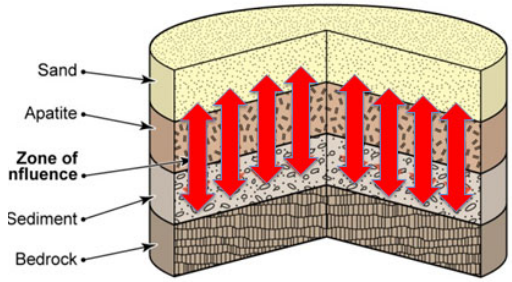
ZOI – zone of influence

# Technical Approach

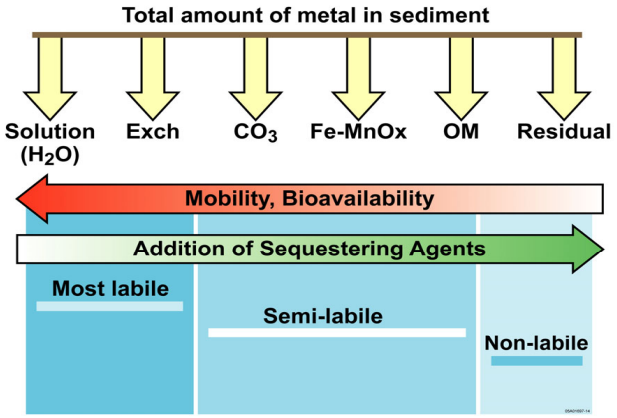
**Hypotheses:**

1) **A Zone of Influence (ZOI) will form in contaminated sediment that is deposited over active caps resulting in chemical changes to the contaminants that will reduce their environmental impact**
2) **The amendments in active caps will sequester contaminants associated with the continued influx of contaminants**



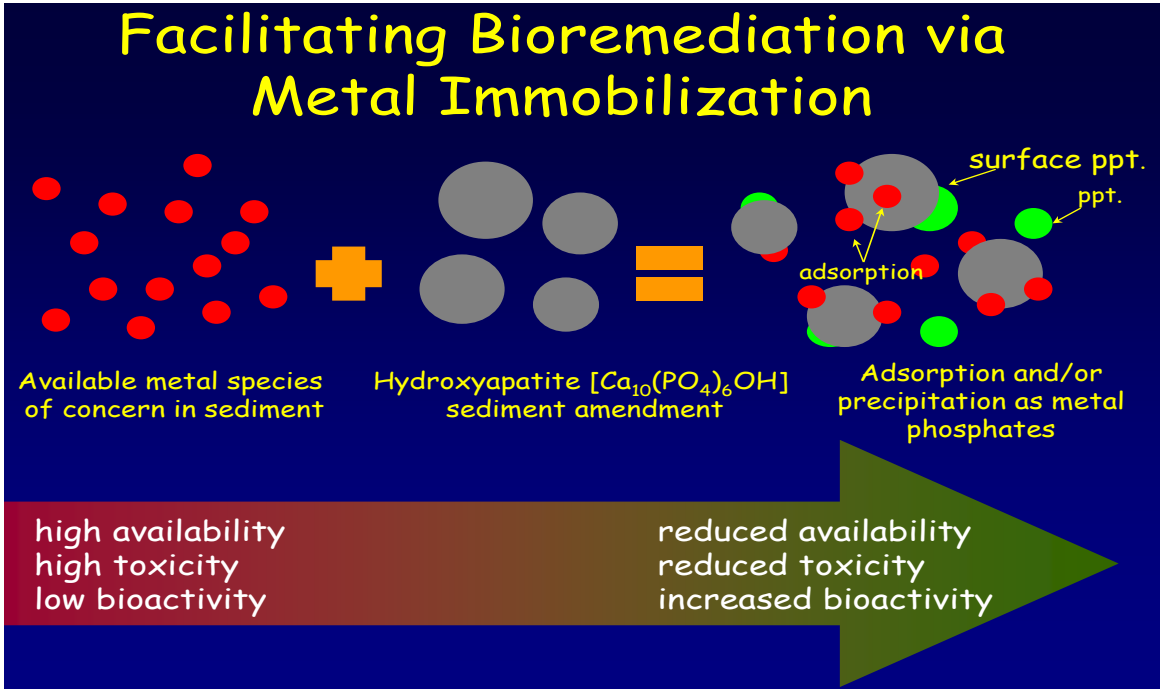

**Active caps remediate existing contaminants in sediments and control/remediate ongoing sources**

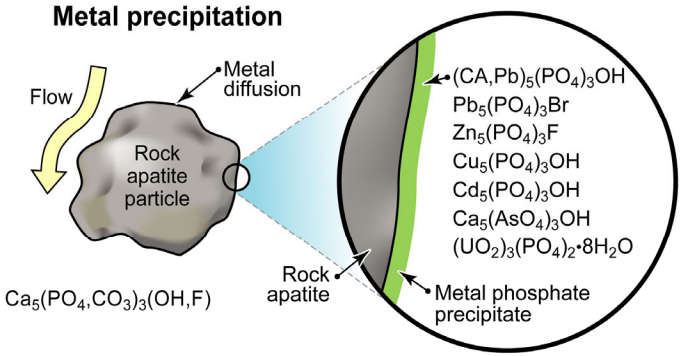


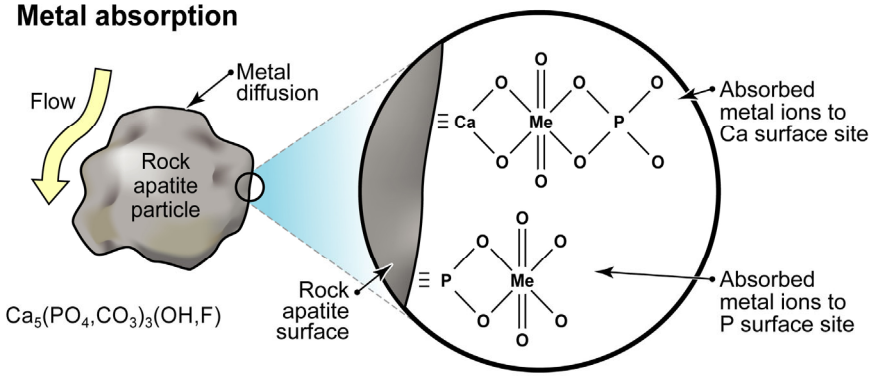
# Remediation via Apatite

## Apatite

- Stable end-products
- Can be placed by existing technology
- Does not affect sediment physical properties
- Can be mixed with other additives
- Low cost, readily available, non-toxic



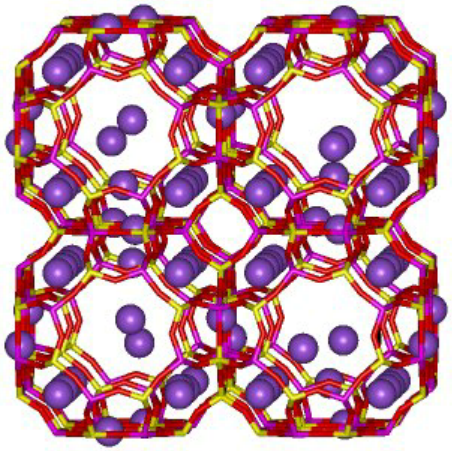



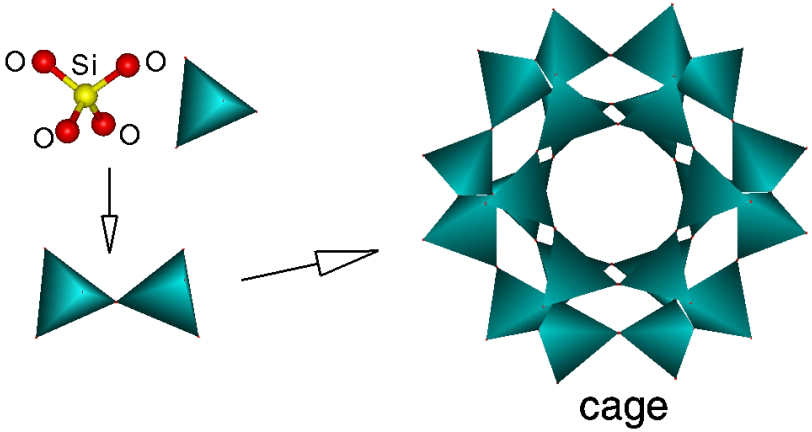
# Sequestering Agents

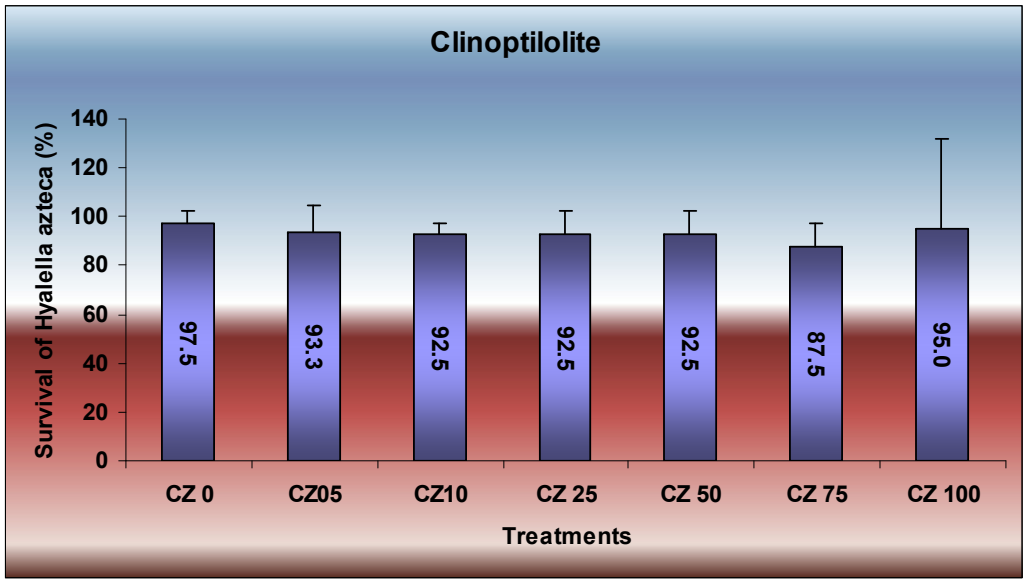
## Zeolites "boiling stones"

- Naturally occurring aluminosilicate minerals
- Have three-dimensional framework with large vacant cages for cations and large molecules
- Clinoptilolite and phillipsite - common zeolites for metal removal
- Clinoptilolite is not toxic to aquatic organisms

## Zeolite Structures



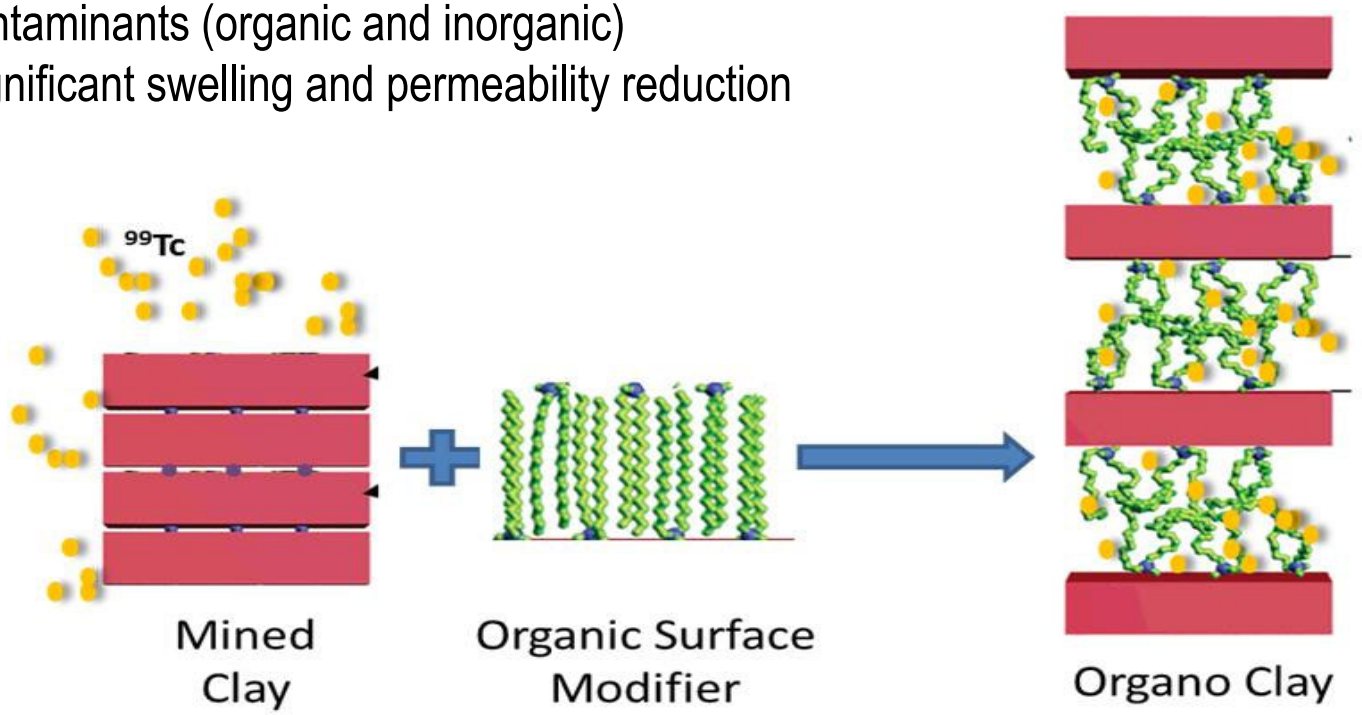


# Sequestering Agents

## Organoclays

- Consist of modified bentonite with organic surface modifiers that increase the surface area of the mineral and create binding sites for $^{129}I$, $^{99}Tc$ and other contaminants (organic and inorganic)
- Significant swelling and permeability reduction



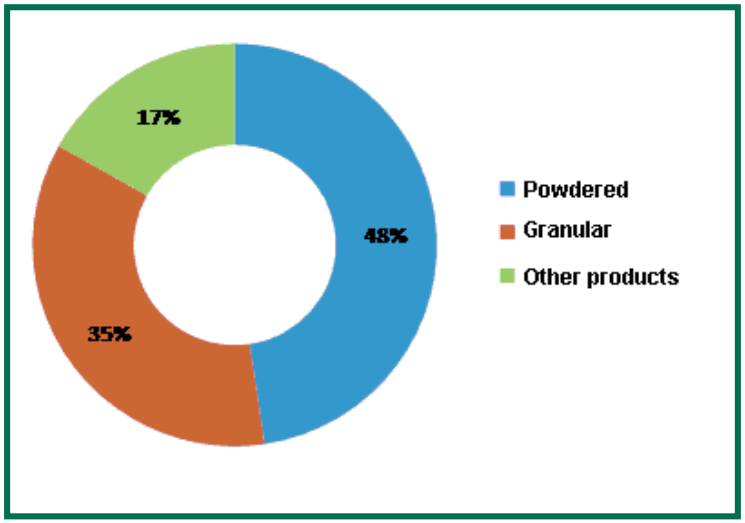

Li, D., Kaplan, D. I., Knox, A. S., Crapse, K. P., and Diprete, D. P. (2014). Aqueous $^{99}Tc$, $^{129}I$ and $^{137}Cs$ removal from contaminated groundwater and sediments using highly effective low-cost sorbents. *Journal of Environmental Radioactivity* **136**, 56-63

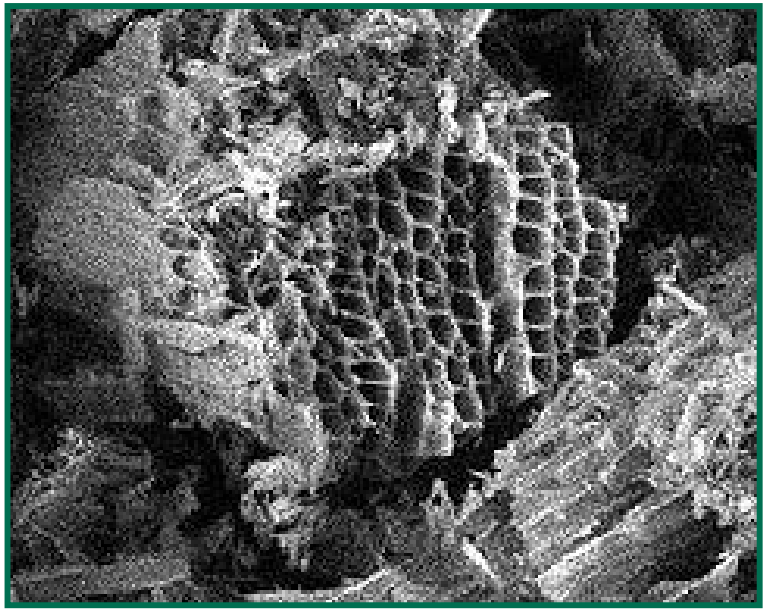
# Sequestering Agents

**Activated carbon (AC)** is particles of carbon that have been treated to increase their surface area and increase their ability to adsorb a wide range of contaminants - activated carbon is particularly good at adsorbing organic compounds.

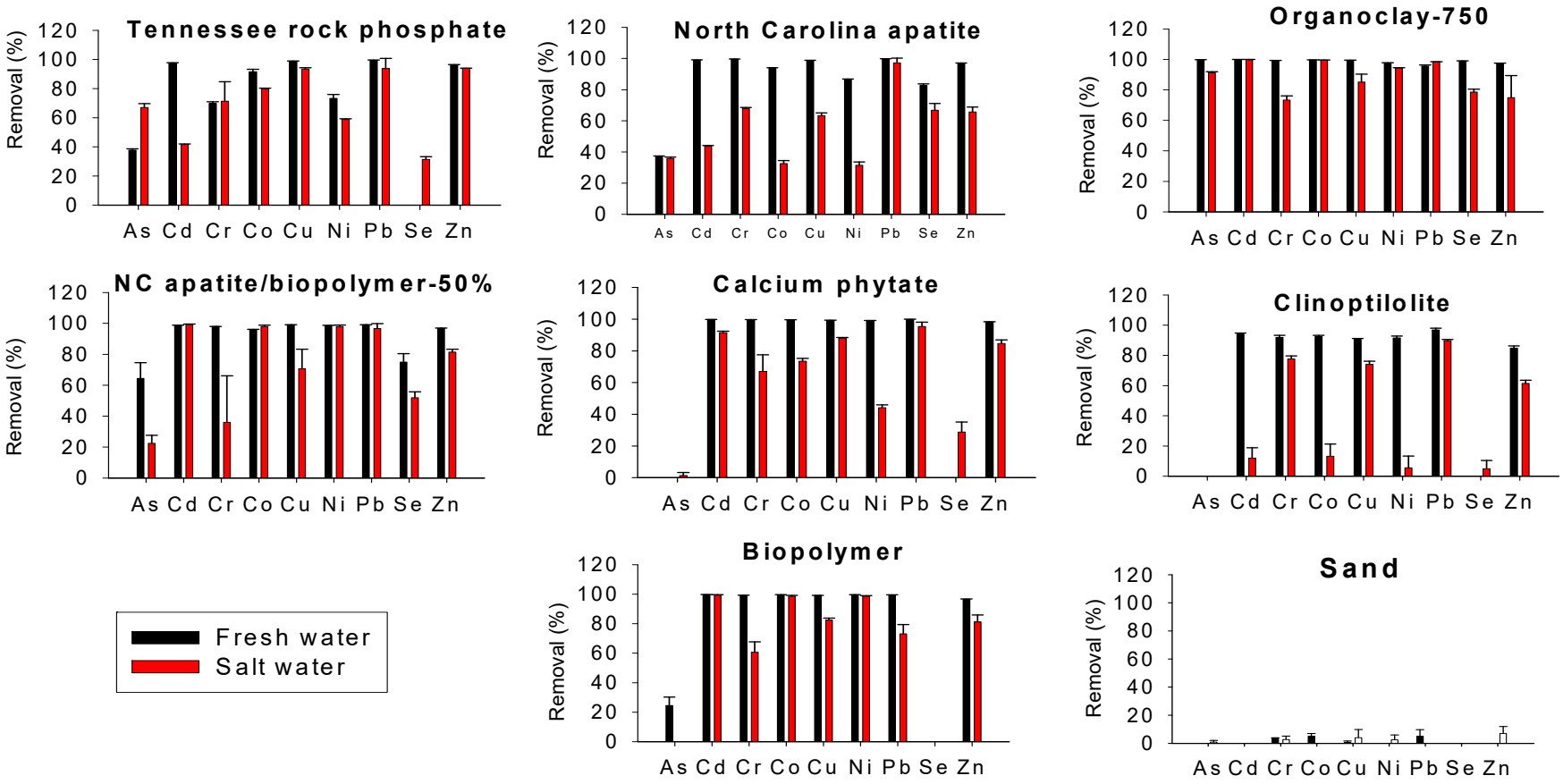
**AC is a highly porous material**

- It has an extremely high surface area for contaminant adsorption
- The equivalent surface area of 1 pound of AC ranges from 60 to 150 acres



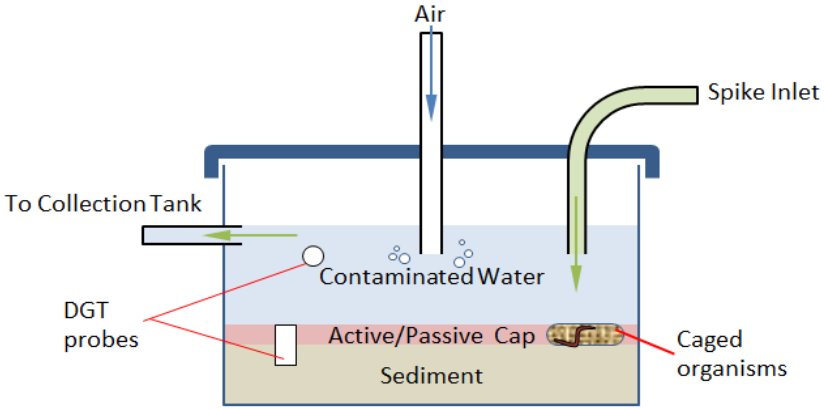



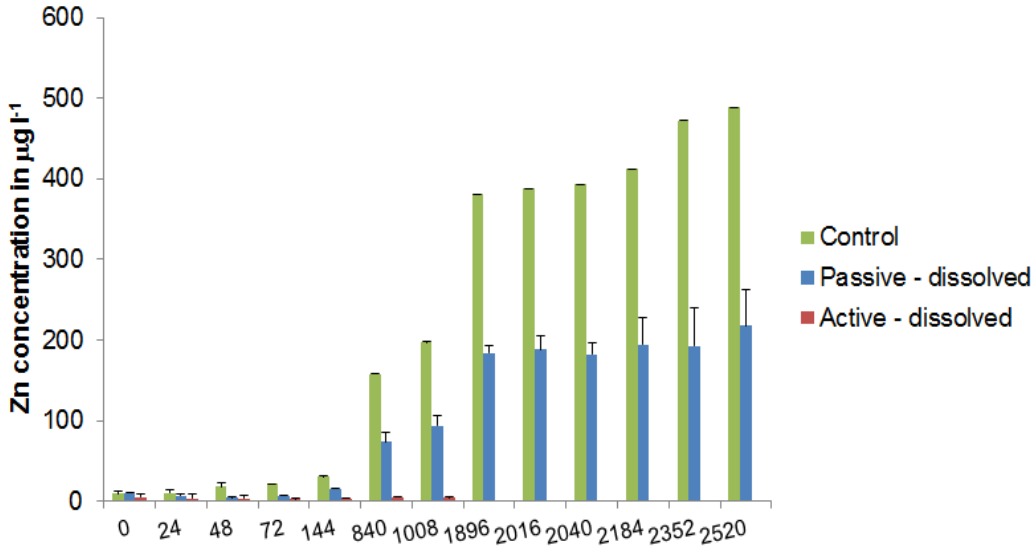
# Removal of Metals

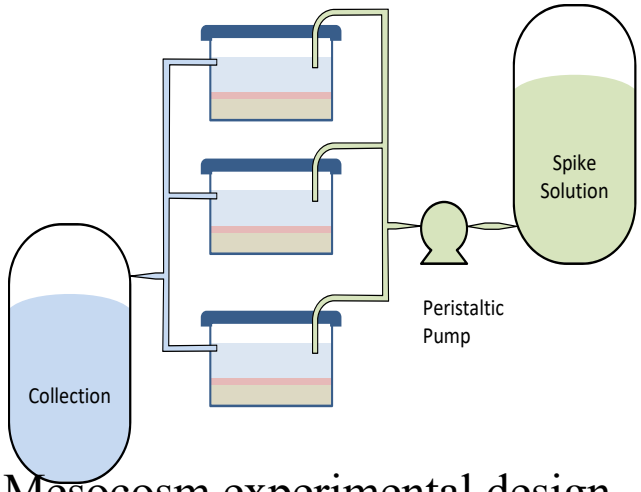



**Comparison of metal removal by amendments in fresh and salt water**

## Average surface water concentrations of dissolved Zn in mesocosms with passive caps (sand), active caps (apatite, activated carbon and mixture of active amendments), and without caps or sediment (control) over a period of 2520 hours.



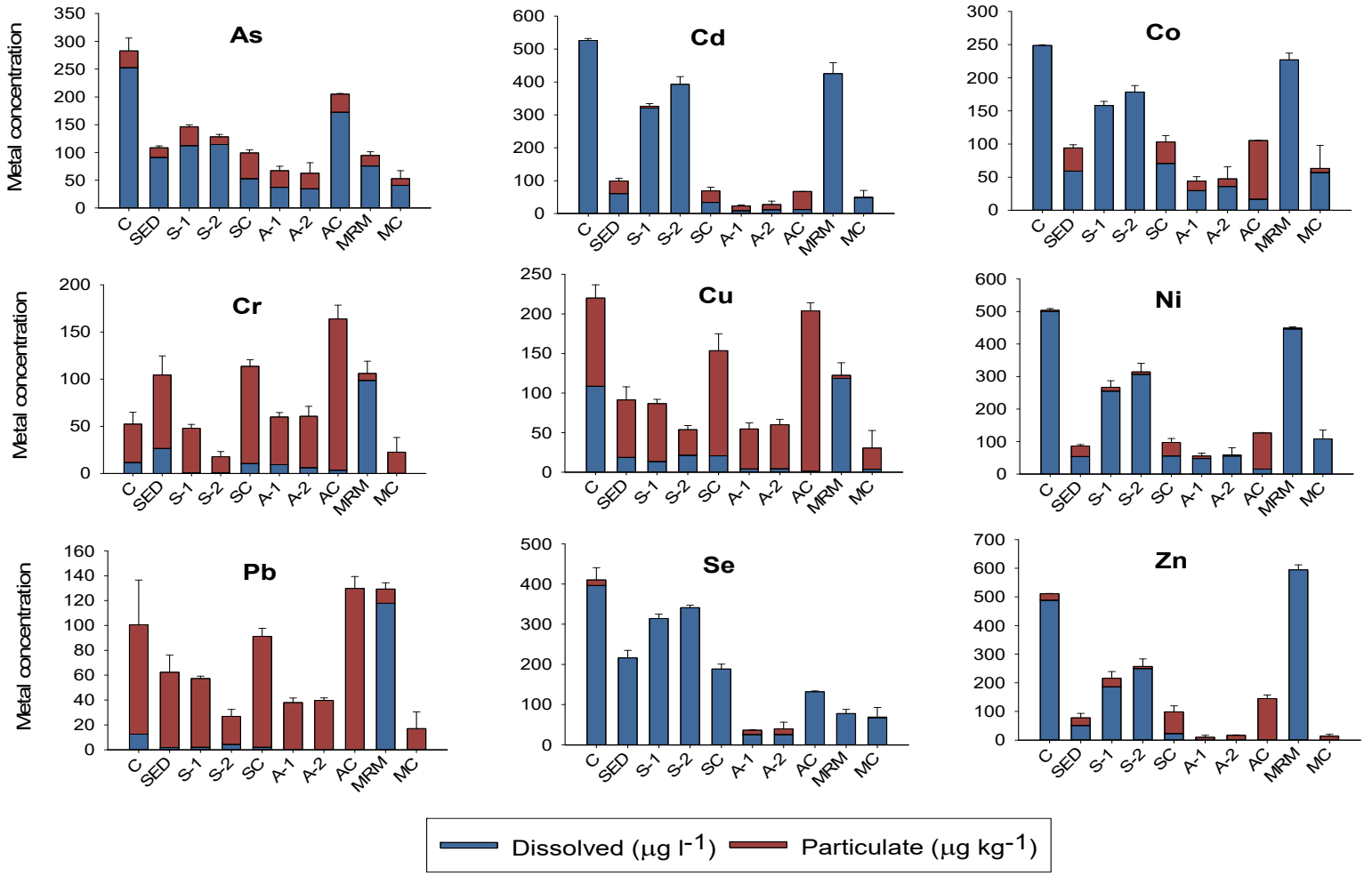




Mesocosm experimental design

Spike solution (C), uncapped sediment (SED), sediment with passive sand caps (S-1: 2.5 cm, S-2: 5 cm), and sediment with several types of active caps (A-1: 2.5 cm apatite , A-2: 5.0 cm apatite, AC: activated carbon, MRM: 2.5 cm organoclay, and MC: 2.5 cm mixture of active amendments)

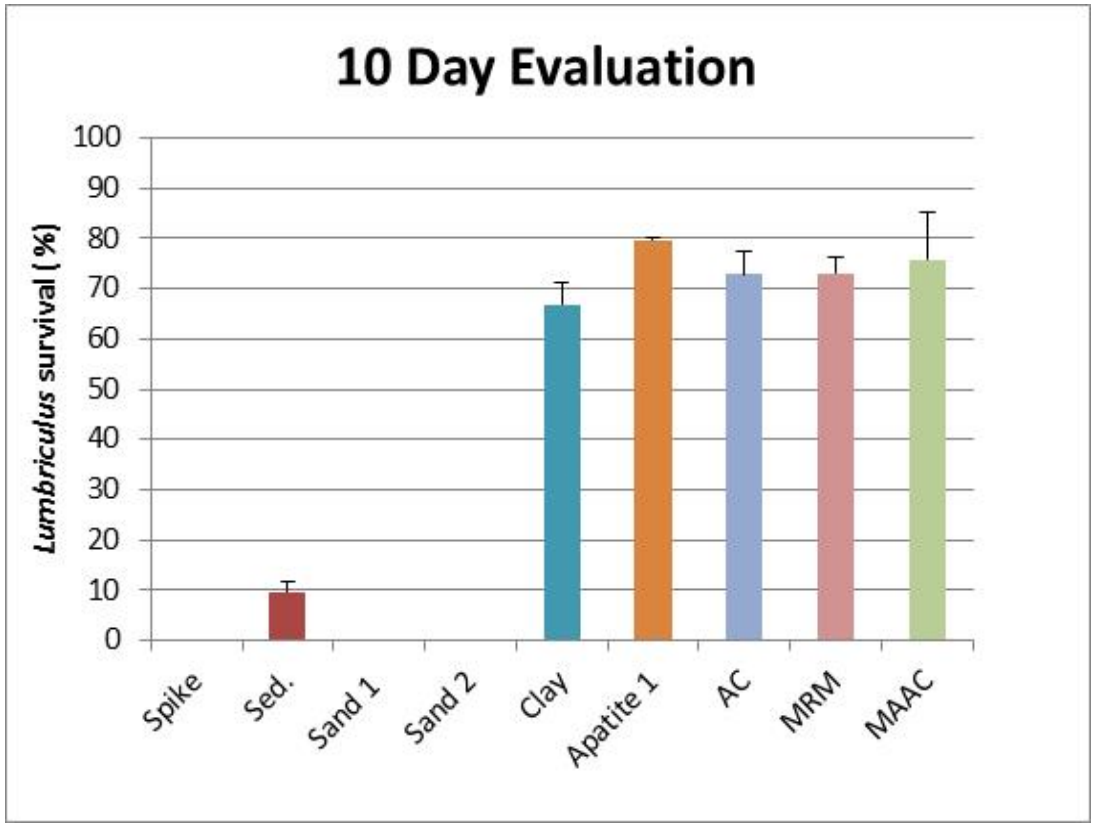
# Average surface water concentration of metals in mesocosms at 2520 hours




Spike solution (C), uncapped sediment (SED), sediment with passive sand caps (S-1: 2.5 cm, S-2: 5 cm), and sediment with several types of active caps (SC: 2.5 cm silty clay, A-1: 2.5 cm apatite , A-2: 5.0 cm apatite, AC: activated carbon, MRM: 2.5 cm organoclay, and MC: 2.5 cm mixture of active amendments) (Knox et al., 2016)

# Effect of Passive and Active Caps on Contaminant Toxicity from Ongoing Sources of Contamination

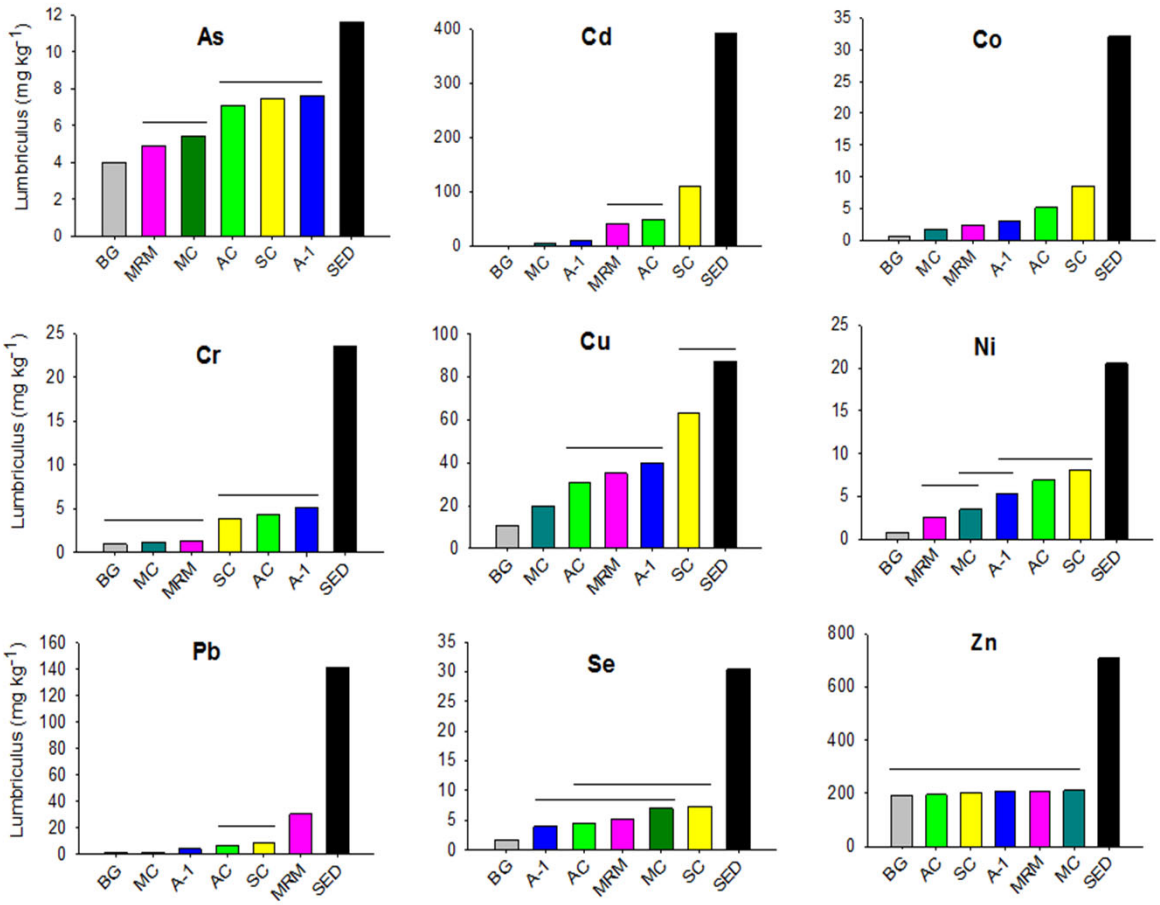
***Lumbriculus variegatus*** were observed for toxicity. Sand caps and spike solution alone (0.5 mg/L of As, Cd, Cr, Cu, Ni, Se, Pb, and Zn) resulted in 100% mortality after 24 hours. However, active caps and clay cap showed minimal toxicity after one, six, and ten days in the presence of spike solution.




Sed. – uncapped sediment, AC – activated carbon, MRM – organoclay, MAAC – Multiple Amendment Active Cap

# Effect of Cap Treatments on Metal Uptake by *Lumbriculus* from Ongoing Sources

## Active caps remediate existing contaminants in sediments and control/remediate ongoing sources

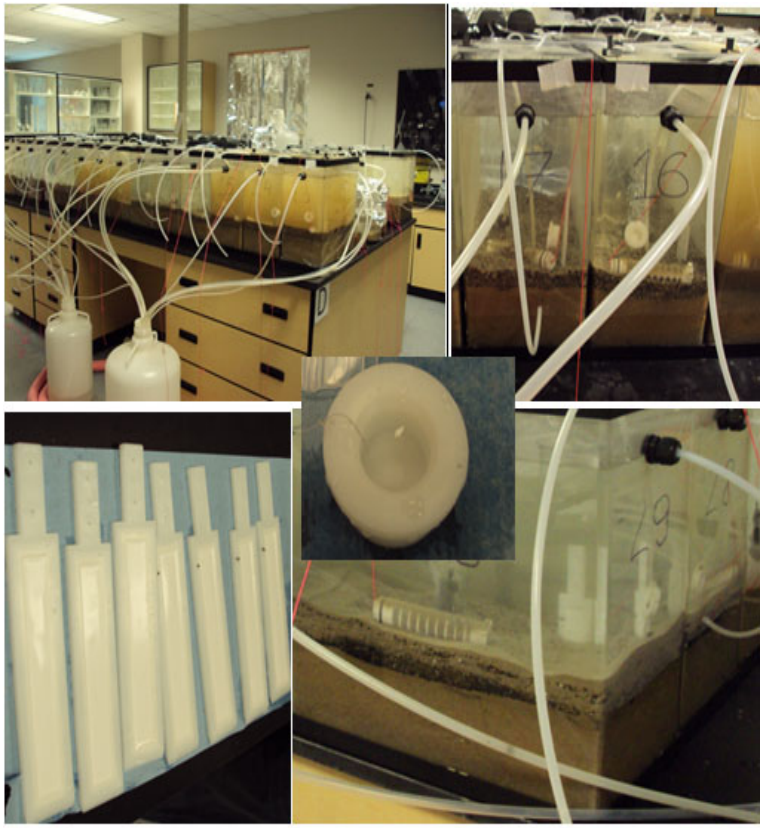



Knox et al., 2016

**Analysis of variance of differences in *Lumbriculus variegatus* metal concentrations (whole body, 10 day exposure)** among sediment treatments (BG =background, AC: activated carbon, SC: silty clay cap, A-1: apatite cap (2.5 cm), MRM: organoclay MRM cap, MC: mixture of active amendments, SED: untreated sediment). Geometric means connected by the same line are not significantly different at $p<0.05$.

# Evaluation of Bioavailable Pool of Metals in Remediated Sediments

- The bioavailable pool of metals in the water and sediment/cap was measured by two types of diffusive gradients in thin films (DGT)
- DGT measurements were compared with metal uptake by caged California black worms (*Lumbriculus variegatus*)



Placement and retrieval of California black worms

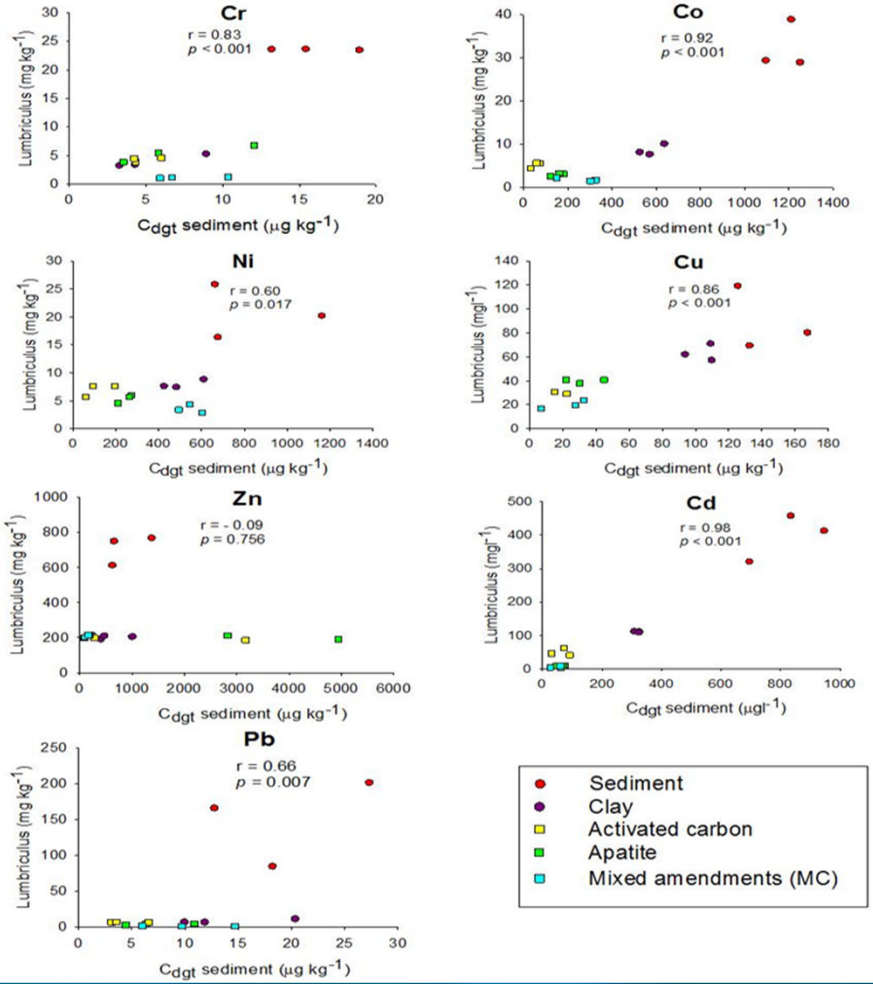
Knox et al., 2016

# Evaluation of Metal Bioavailable Pool in Contaminated sediment Treated with Passive and Active Caps

## Metal concentrations in both *Lumbriculus* and sediment/cap were lowest in apatite, mixed amendment, and activated carbon treatments

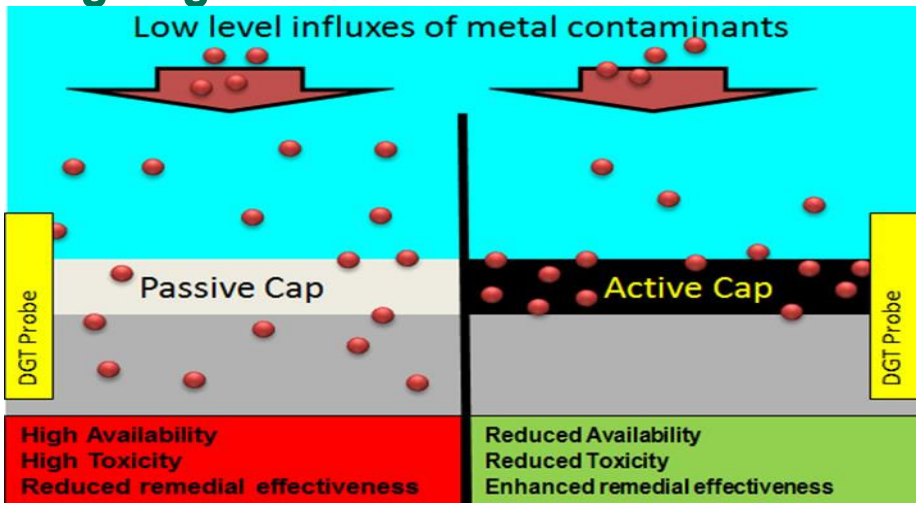
- Correlations between *Lumbriculus and* sediment concentrations measured by DGT sediment probes were strong, confirming the effectiveness of active caps

Pearson correlations between metal concentrations in *Lumbriculus* (ten day test) and metal concentrations in the top 2.5 cm of sediment or cap measured by diffusive gradient in thin films (DGT) sediment probes were strong (as high as 0.98) and significant ($p<0.05$) for almost all tested metals.



# Active Capping Advantages

- Can achieve greater risk reduction more quickly
- Creates less short-term risk
- Can be implemented more quickly and economically
- Does not require staging, handling, and treatment of removed sediment
- Can facilitate habitat restoration by using an eco-friendly surface layer
- **Active caps remediate existing contaminants in sediments and control/remediate ongoing sources**

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Environmental impact of ongoing sources of metal contamination on remediated sediments

Anna Sophia Knox [a,*], Michael H. Paller [a], Charles E. Milliken [a], Todd M. Redder [b], John R. Wolfe [b], John Seaman [c]

# Final Key Points

- All remediation technologies have their advantages and disadvantages
- Selection of remedial action should consider the risk from ongoing sources of contaminants and site characteristics
- Combinations of approaches and technologies that complement and reinforce each other are good options for remediating and managing contaminated sediments

## Trace Elements in Waterlogged Soils and Sediments

Editors:
The back cover description that will be placed here will be routed separately from the copywriting department. You will have a chance to review and make corrections at that time.

- The book is composed of three parts. "Understanding, processes, and needs," provides fundamental knowledge concerning trace element geochemistry in waterlogged soils and sediments.
- The second part of the book, "Bioavailability (chapters 11 to 16)," provides detailed information on the bioavailability of trace elements in the aquatic and semi-aquatic ecosystems
- The third part of the book, "Remediation" (chapters 17 and 18), discusses the remediation of metal contaminated sediments.

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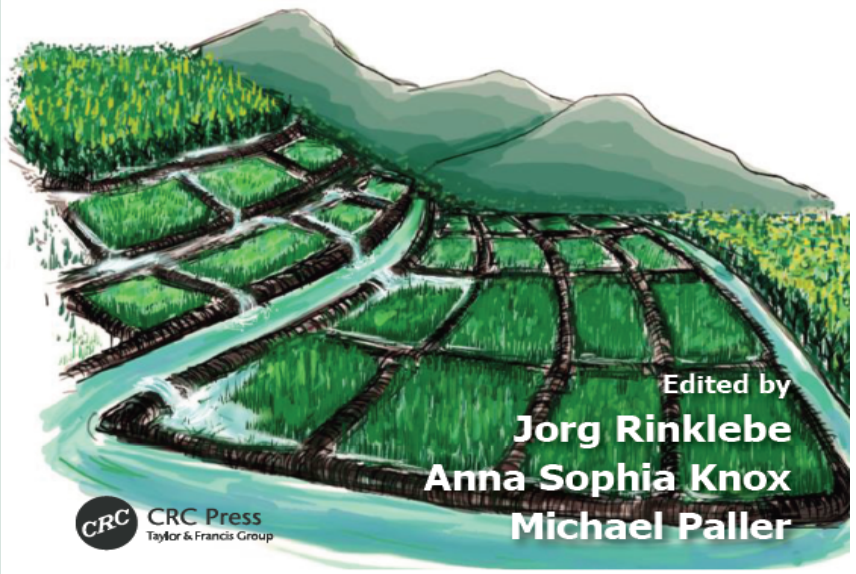


Rinklebe, J., A.S. Knox, and M.H. Paller, editors. 2016. Trace Elements in Waterlogged Soils and Sediments. CRC Press, Boca Raton, FL.