Effect of Aging on Contaminant Bioavailability

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“Priority” Organic Pollutants (HOCs)

- ✓ Chlorinated pesticides
- ✓ PCBs/PBDEs
- ✓ PAHs
- ✓ Dioxins

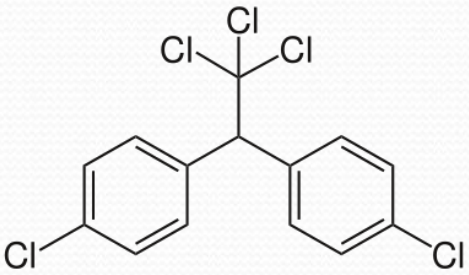
FACT 1:
Strong sorption

- ✓ Chlorinated pesticides
- ✓ PCBs/PBDEs
- ✓ PAHs
- ✓ Dioxins

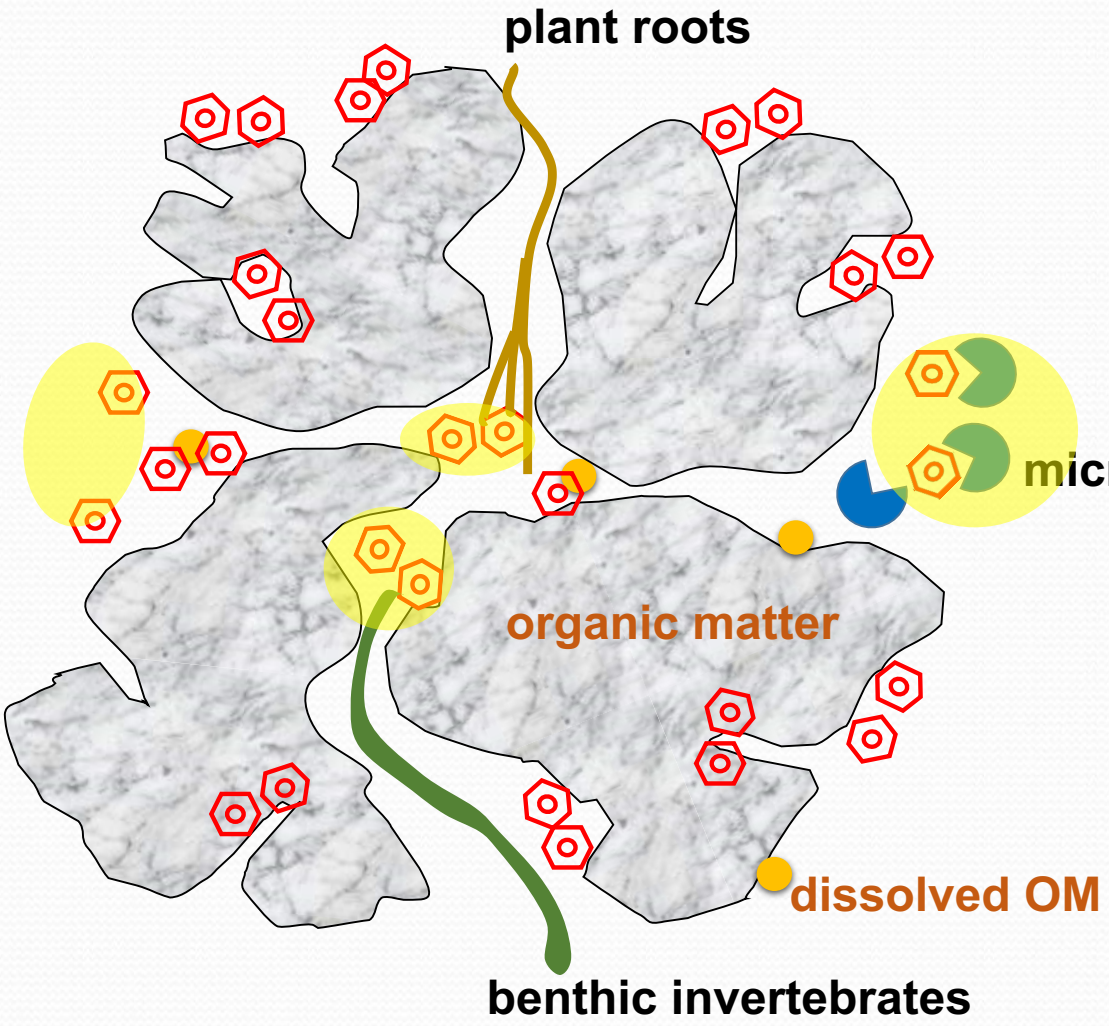
FACT 2:
Aged

DDT

- Paul Hermann Müller, Nobel Prize in Physiology or Medicine, 1948
- During World War II, control malaria and typhus among civilians and troops
- Agricultural insecticide since 1945
- Banned in **1972** in the U.S.

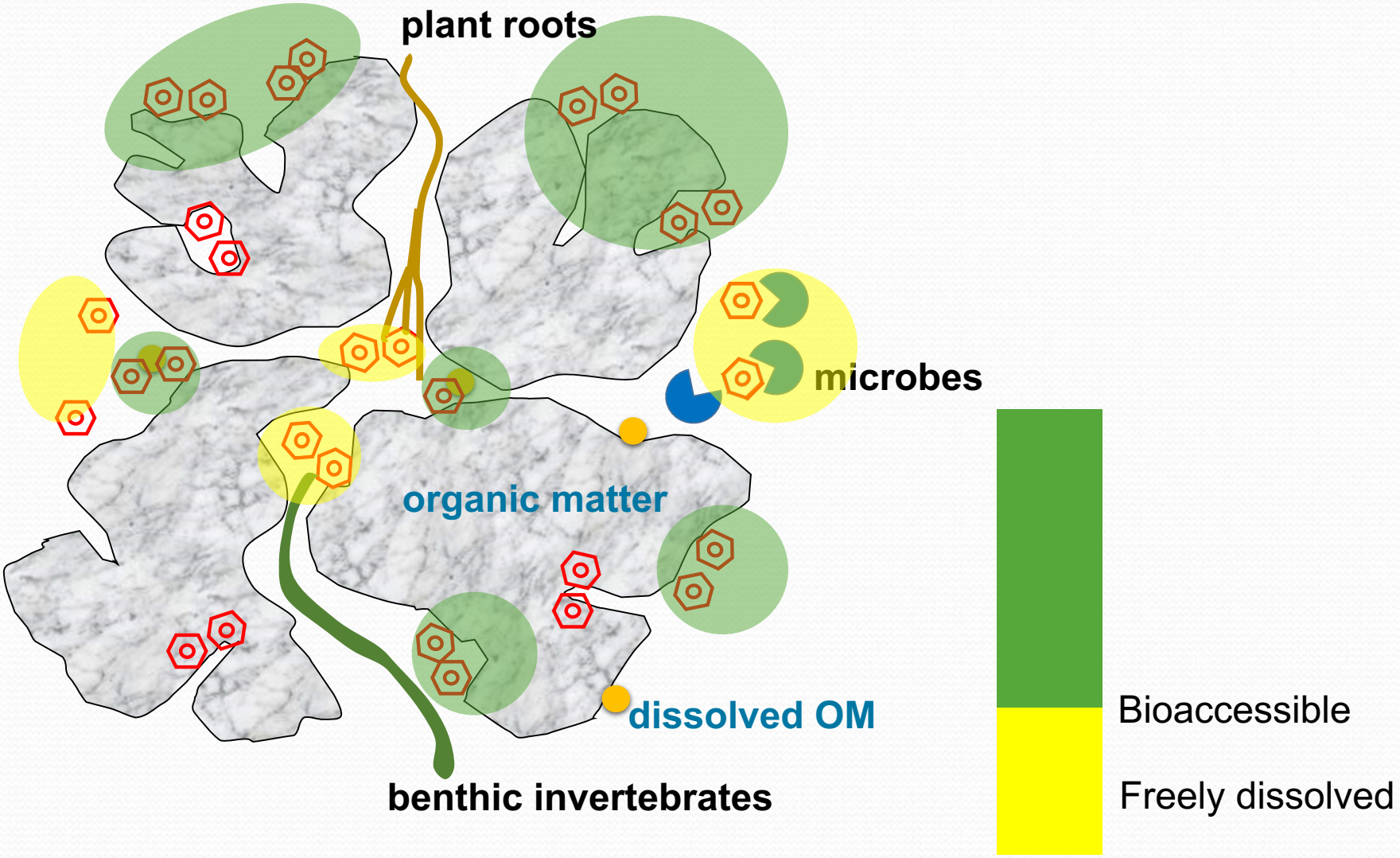


Aging and Bioavailability

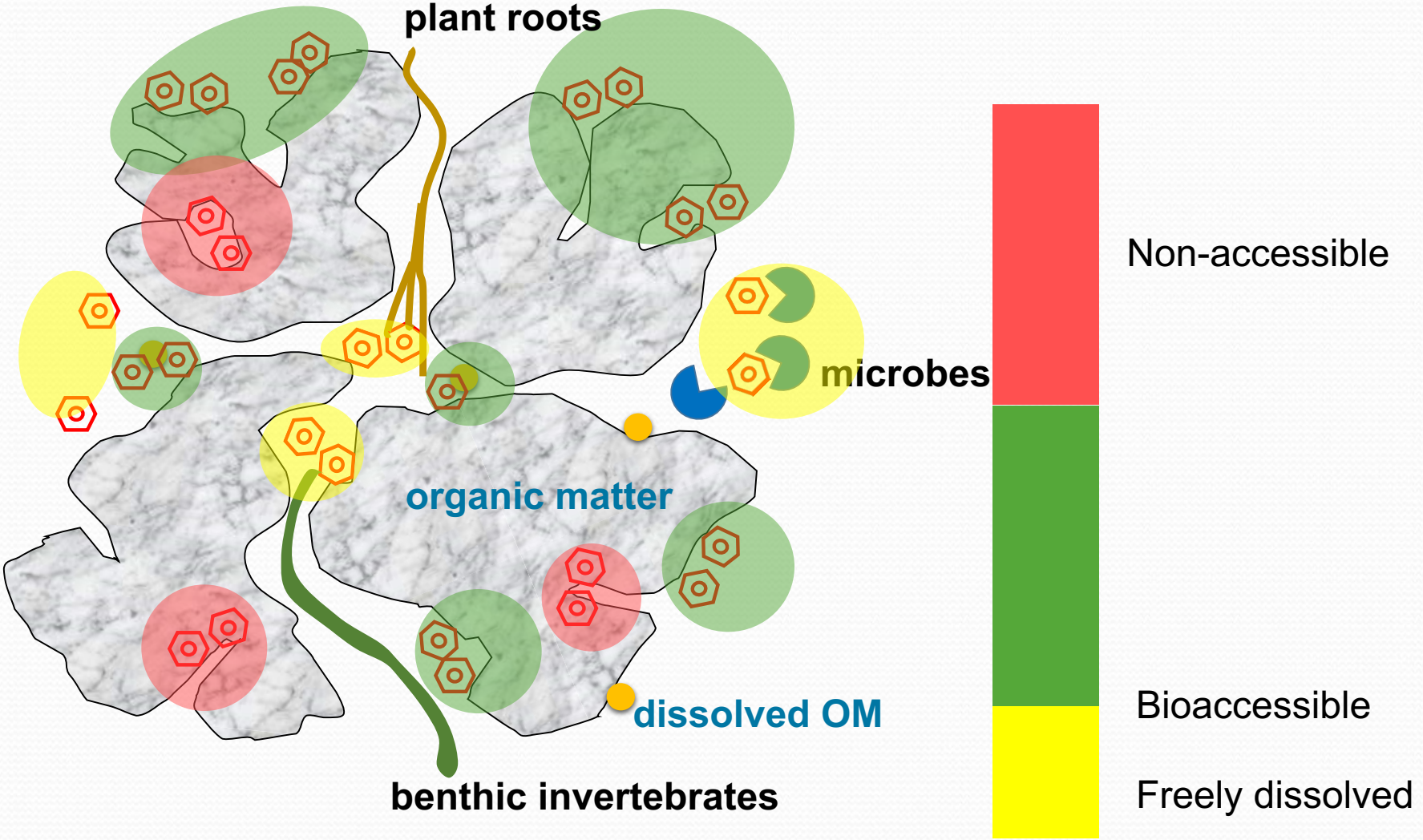


Freely dissolved

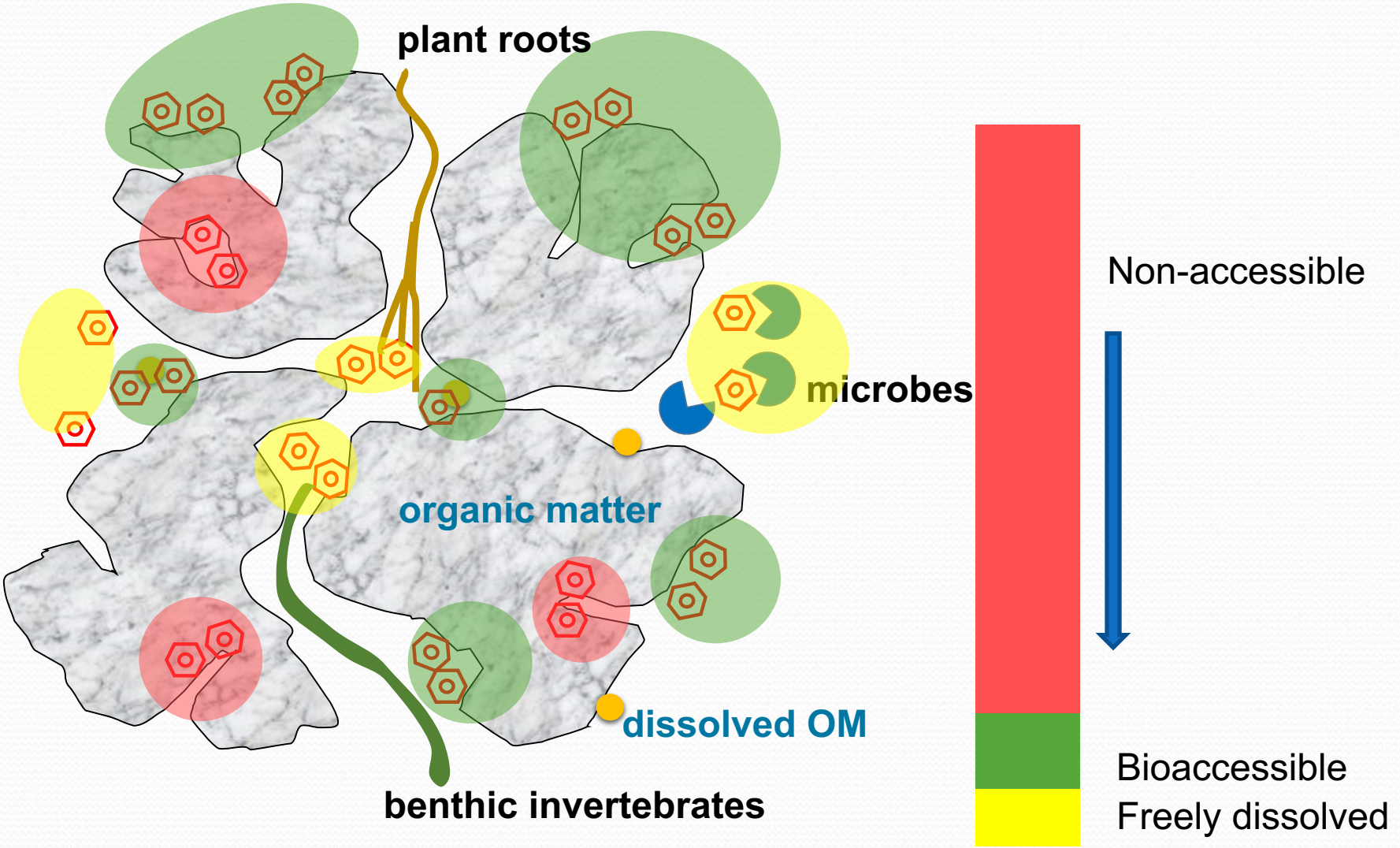
Aging and Bioavailability



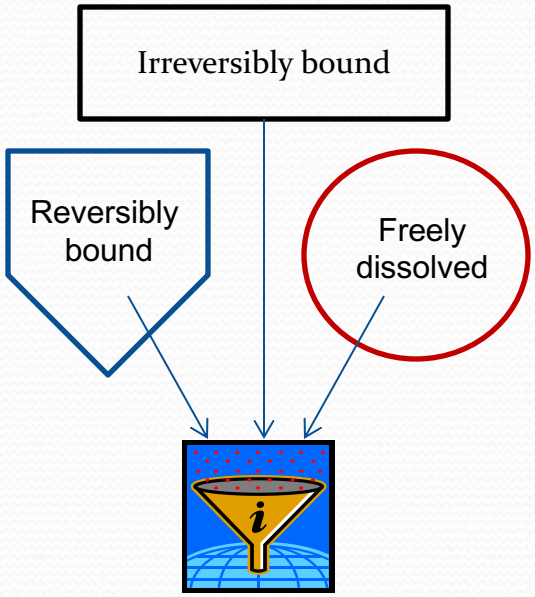
Aging and Bioavailability



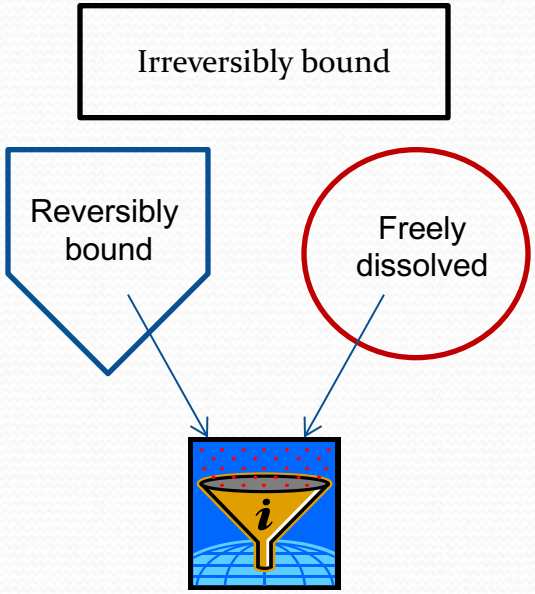
Aging and Bioavailability



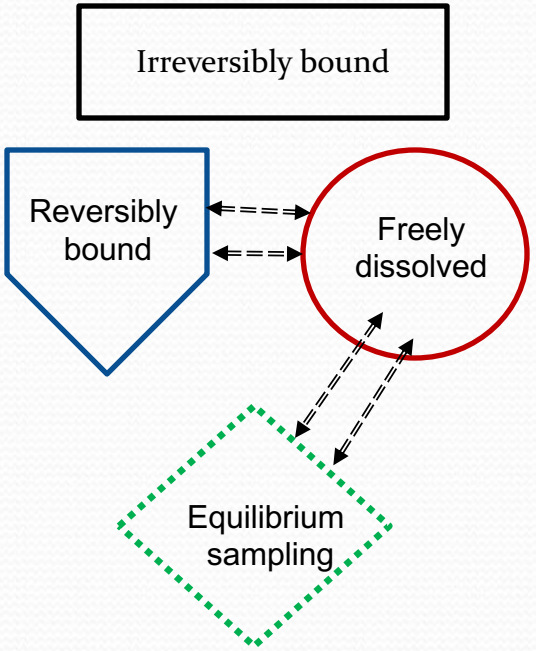
Measuring Bioavailability



Total concentration



Bioaccessibility



C_{free}

Reichenberg and Mayer, ET&C, 2006, 1239-1245

Bioavailability

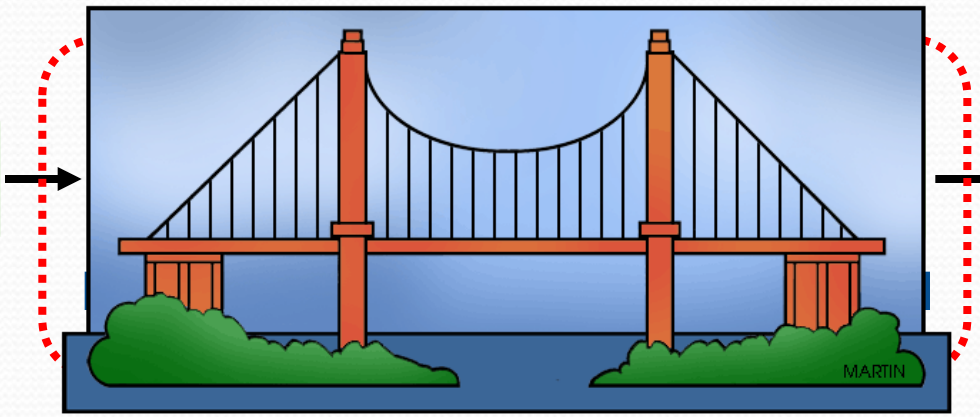
Freely dissolved concentration C_{free}

The potential for a chemical to undergo spontaneous processes, e.g., diffusion and partitioning

Bioaccessibility

The actual amount of a chemical that is or may become available within a given time and under given conditions

Total concentration



Risk

Reichenberg and Mayer, 2006, Environ. Toxicol. Chem. 25, 1239-1245

Bioavailability Methods

Bioaccessibility

- Partial desorption
  - Weak acid extraction
  - Mild solvent extraction
  - Gut fluid extraction
  - Cyclodextrin extraction
  - Tenax adsorption extraction
  - Isotope dilution method (IDM)

C_{free}

- Passive samplers
  - DGT
  - Polyethylene devices (PEDs)
  - Semi-permeable membrane devices (SPMDs)
  - Polyoxymethylene (POM)
  - Solid phase micro-extraction (SPME)

Study I. Aged POPs at the Palos Verdes Superfund Site

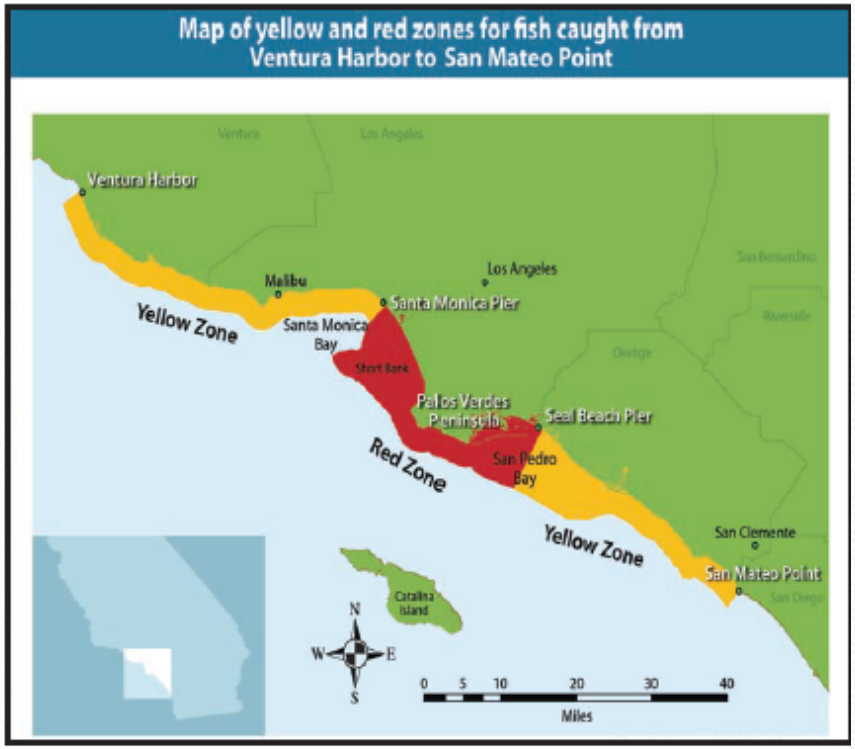
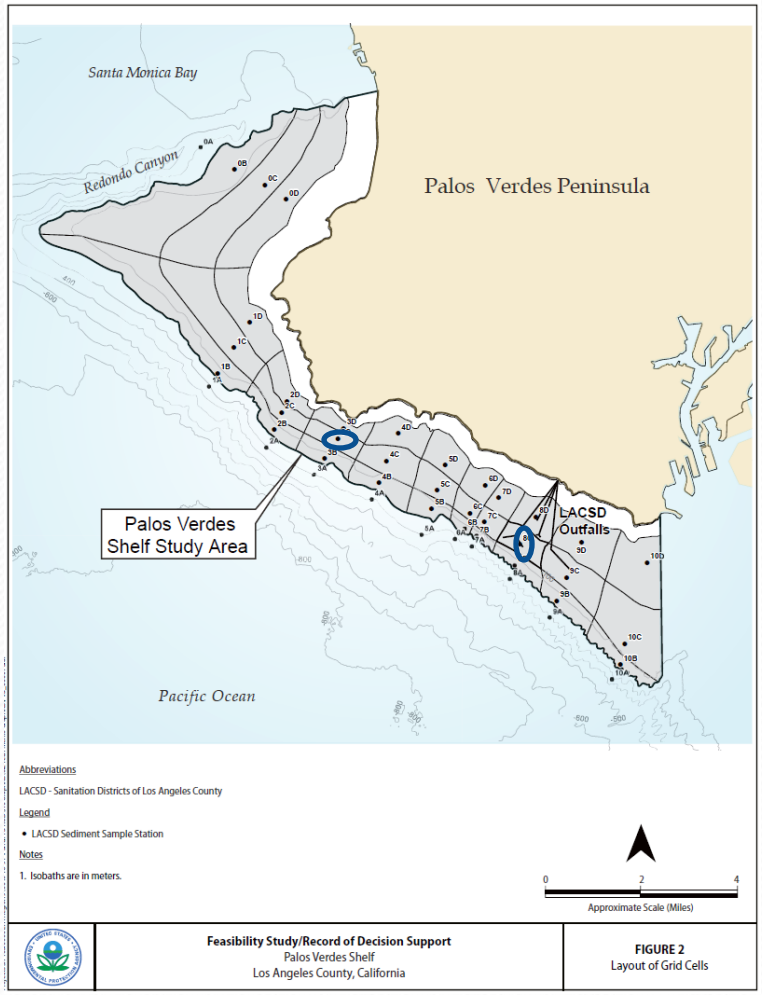


Figure 2-2: OEHHA Health Advisory

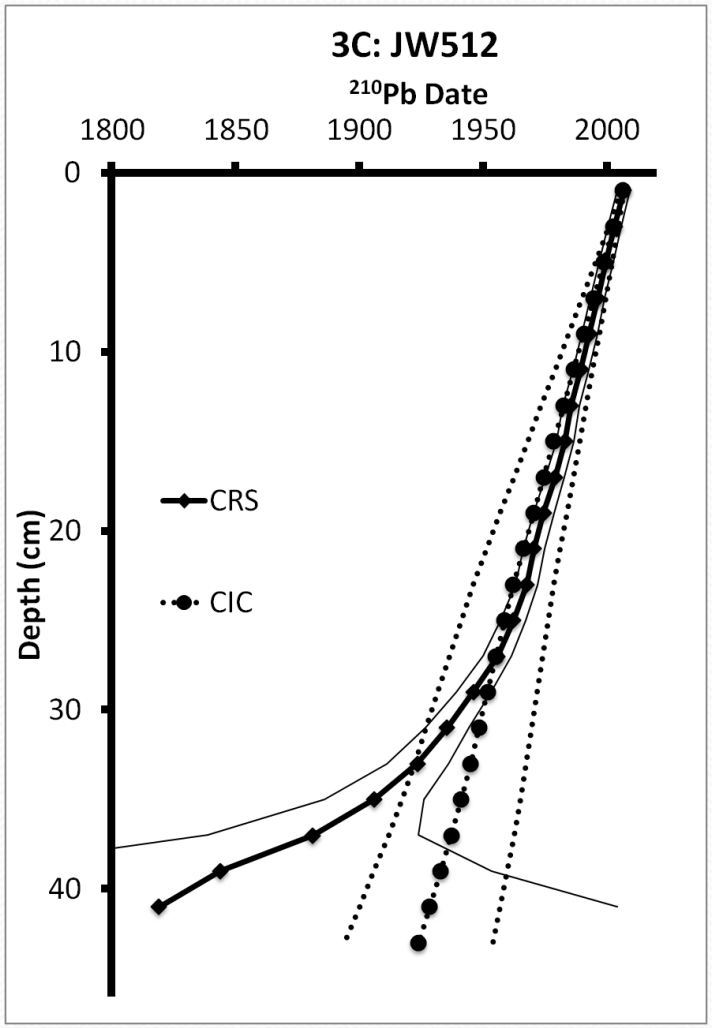
US EPA, 2010

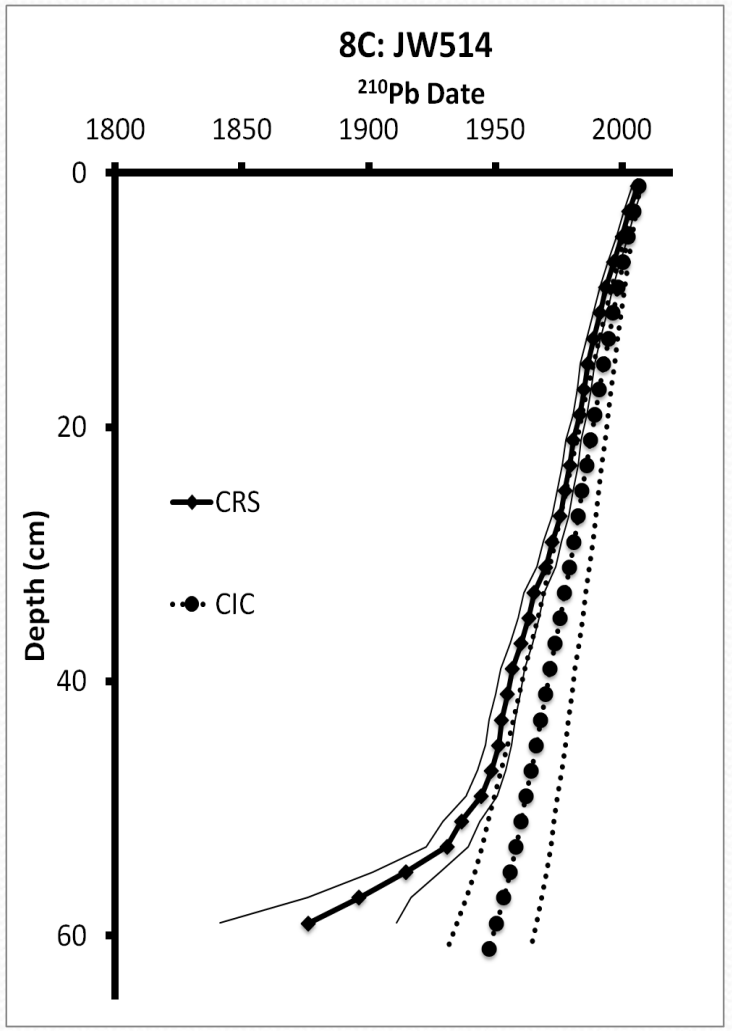
A Sediment Core Experiment

- **Hypothesis:**
  - Contaminant aging has resulted in reduced bioavailability.

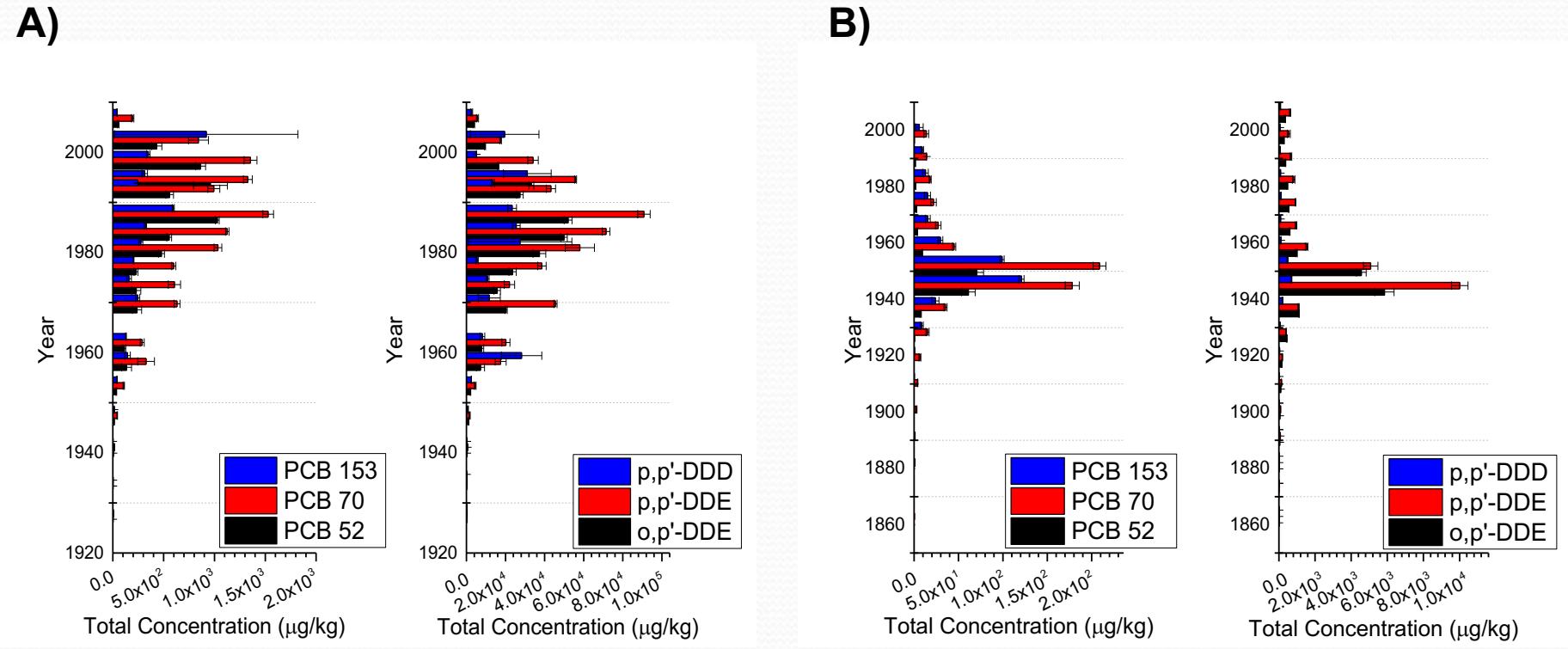


^{210}Pb Dating



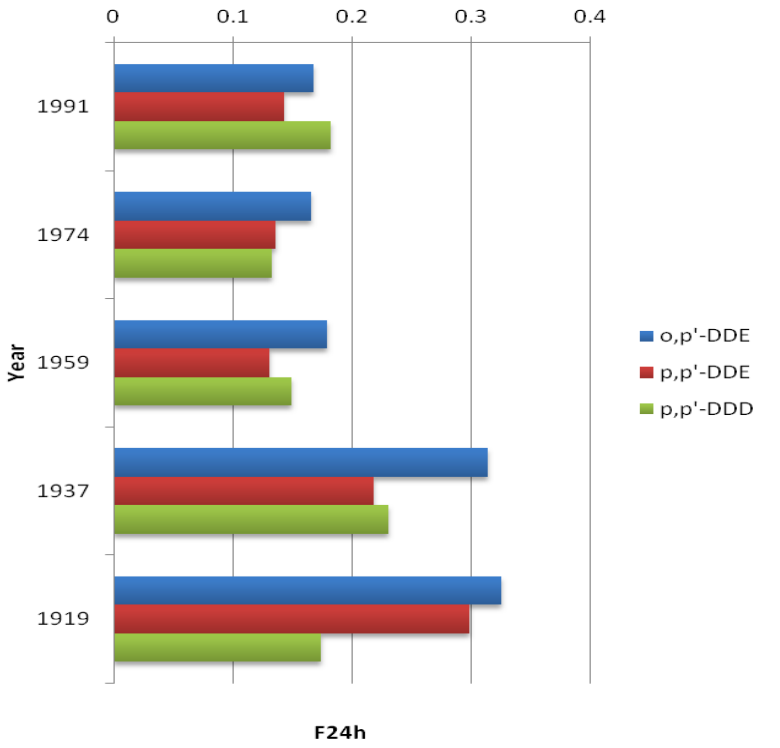


Total Concentrations

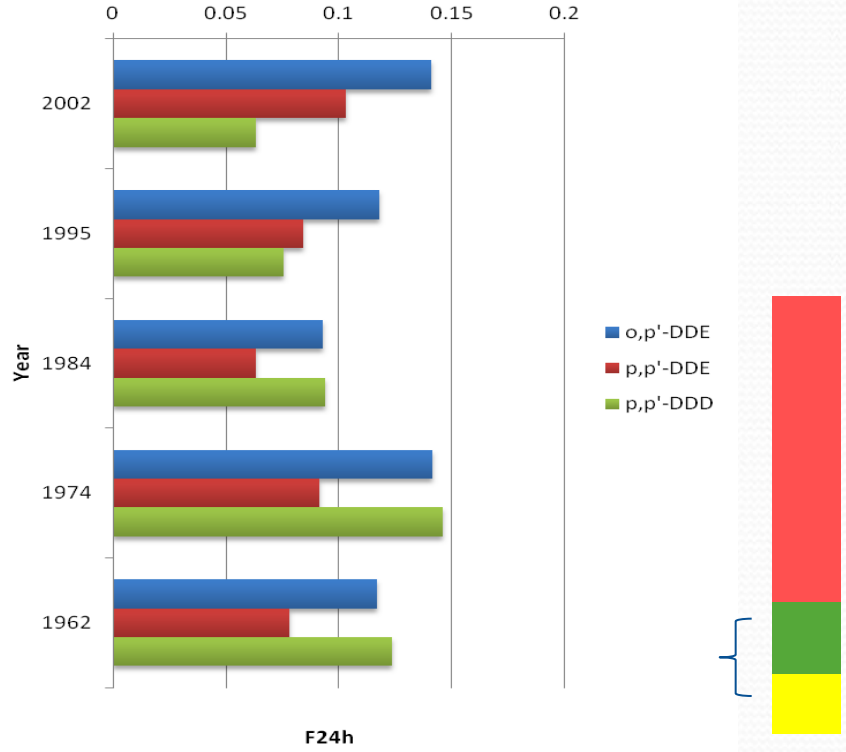


Total concentration profiles of PCBs and DDTs in (**A**) 8C and (B) 3C cores in µg/kg dry weight (d.w.) of sediment.

Tenax Results



3C



8C

Conclusions I

Environmental Pollution 245 (2019) 582–589

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Effect of aging on bioaccessibility of DDTs and PCBs in marine sediment☆

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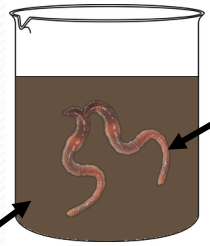
- DDT residues were extensivel
- Due to aging, only a very sma sediment was “bioaccessible”.
- DDTs in surface sediment also showed very low bioaccessibility.
  - DDT residues were “aged” elsewhere before deposition onto the ocean floor
- Implications:
  - Risks much lower than expected from total concentration
  - EPA decided to use MNA (“monitored natural attenuation”) instead of capping.

Study II. A Direct Method to Test Aging

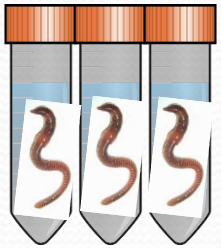
Isotope-labeled HOCs



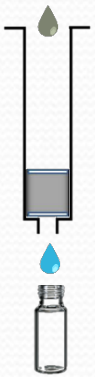
Eisenia fetida

Historically-contaminated soil

Bioaccumulation assay



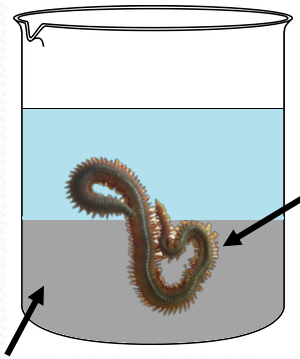
ultrasonic extraction



SPE purification

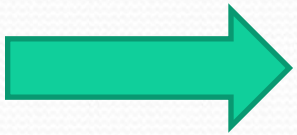
Bioaccumulation assay

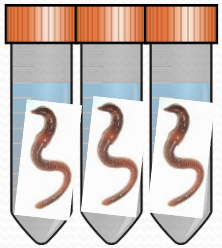
Isotope-labeled HOCs



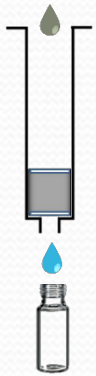
Nereis virens

Historically-contaminated marine sediment





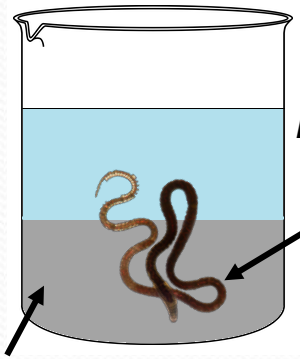
ultrasonic extraction



SPE purification

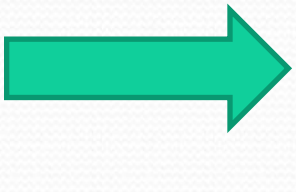
Bioaccumulation assay

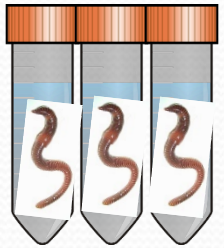
Isotope-labeled HOCs



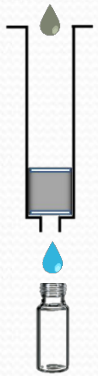
Lumbriculus variegatus

Historically-contaminated freshwater sediment



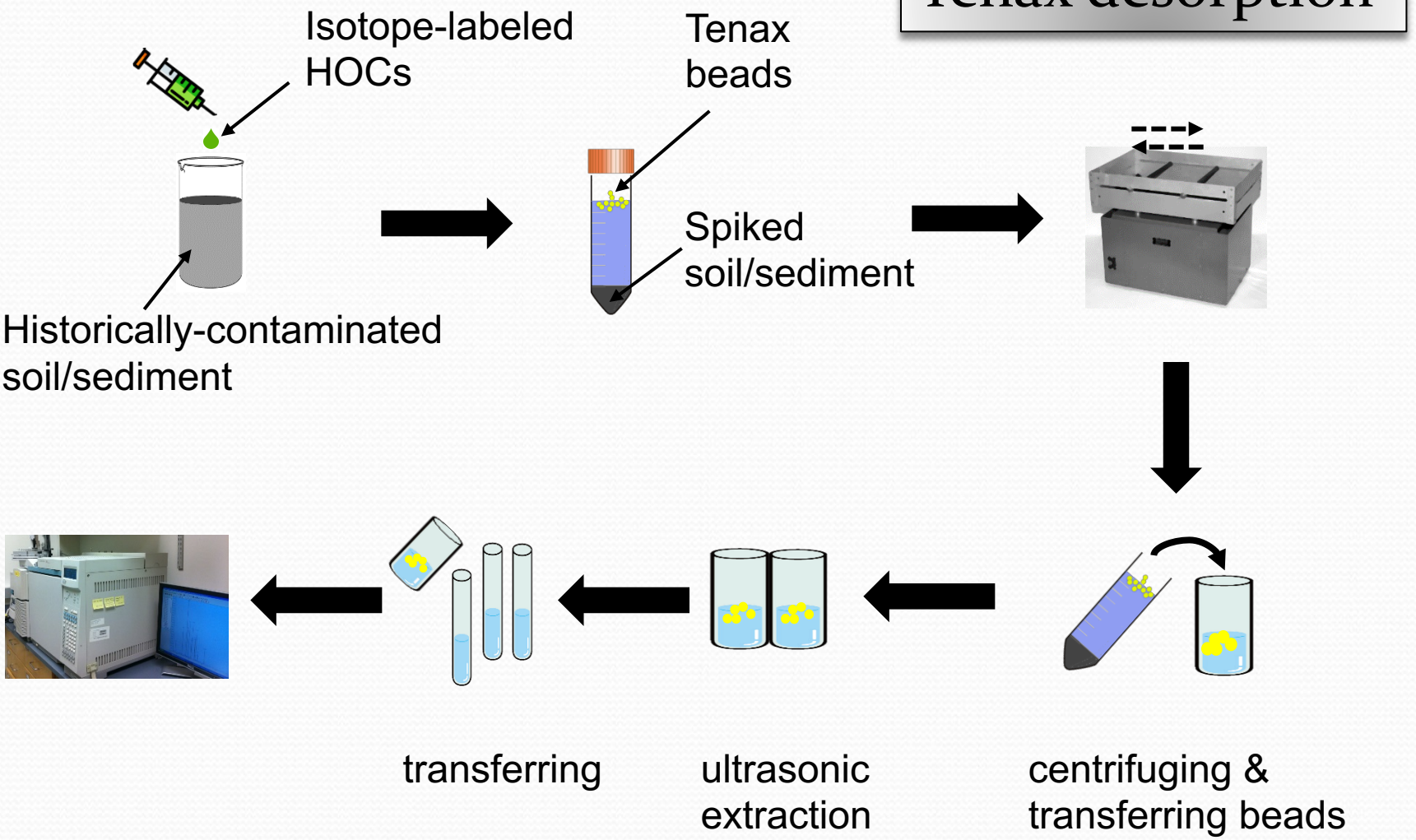


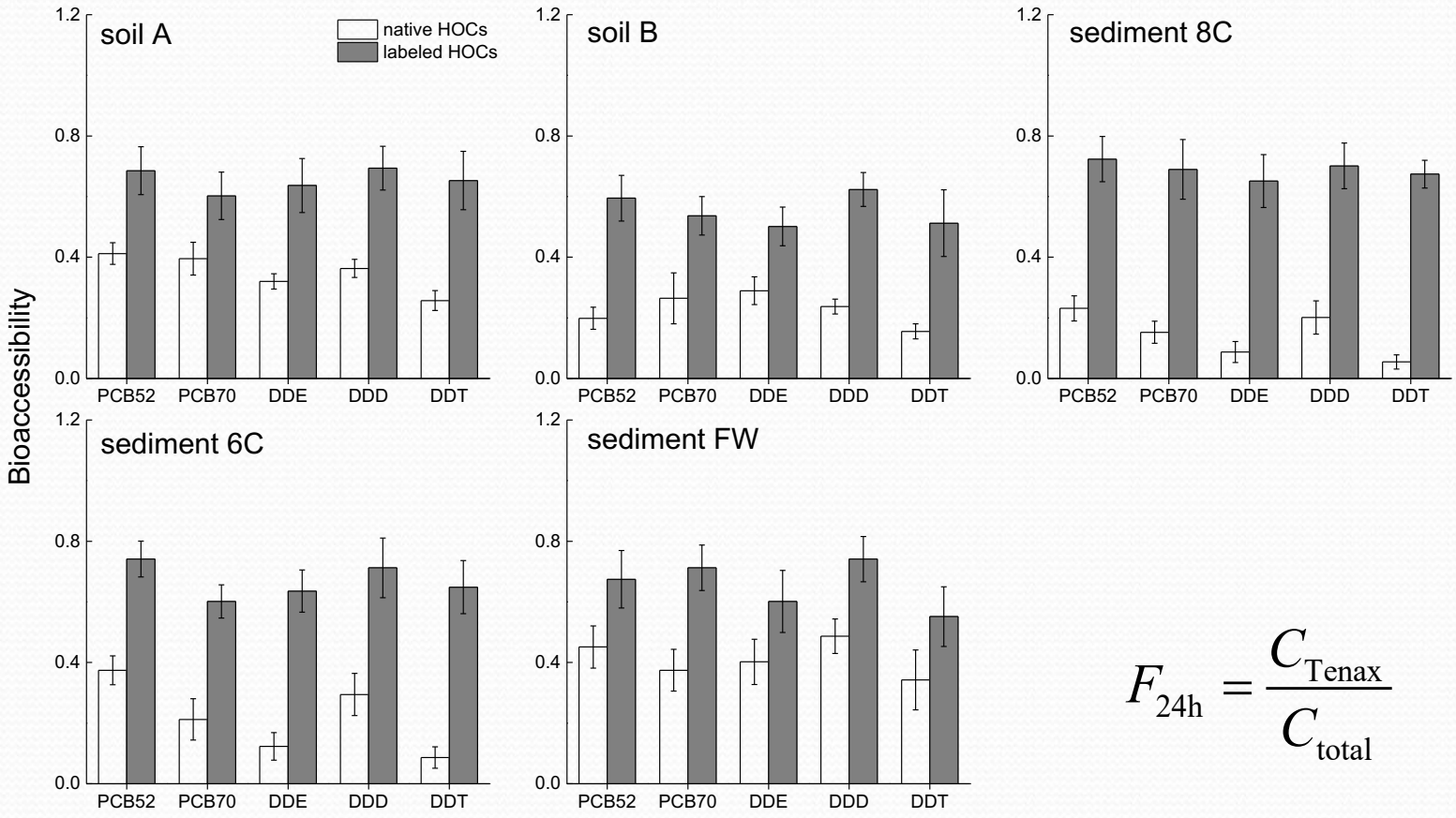
ultrasonic extraction



SPE purification

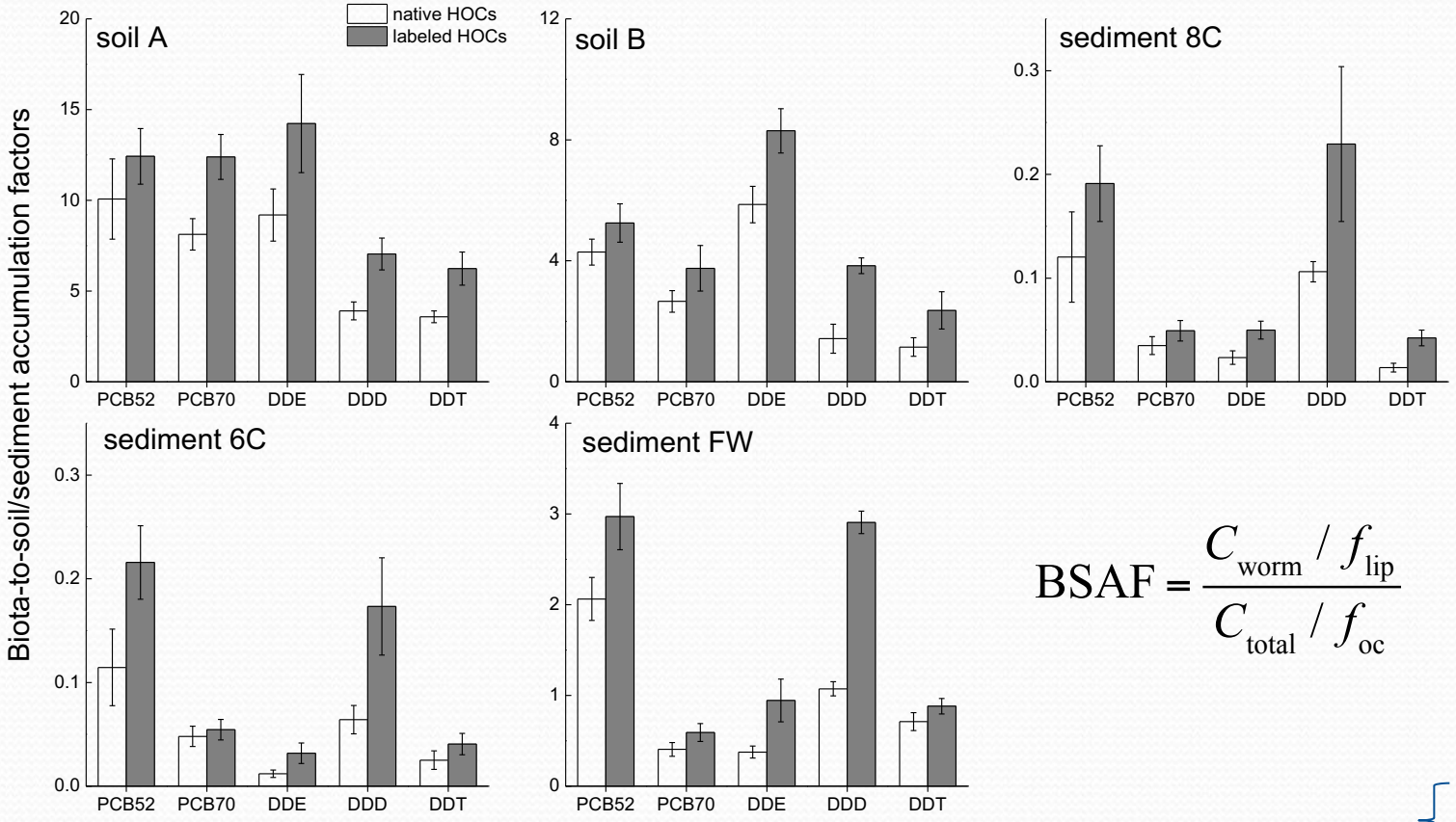
Tenax desorption





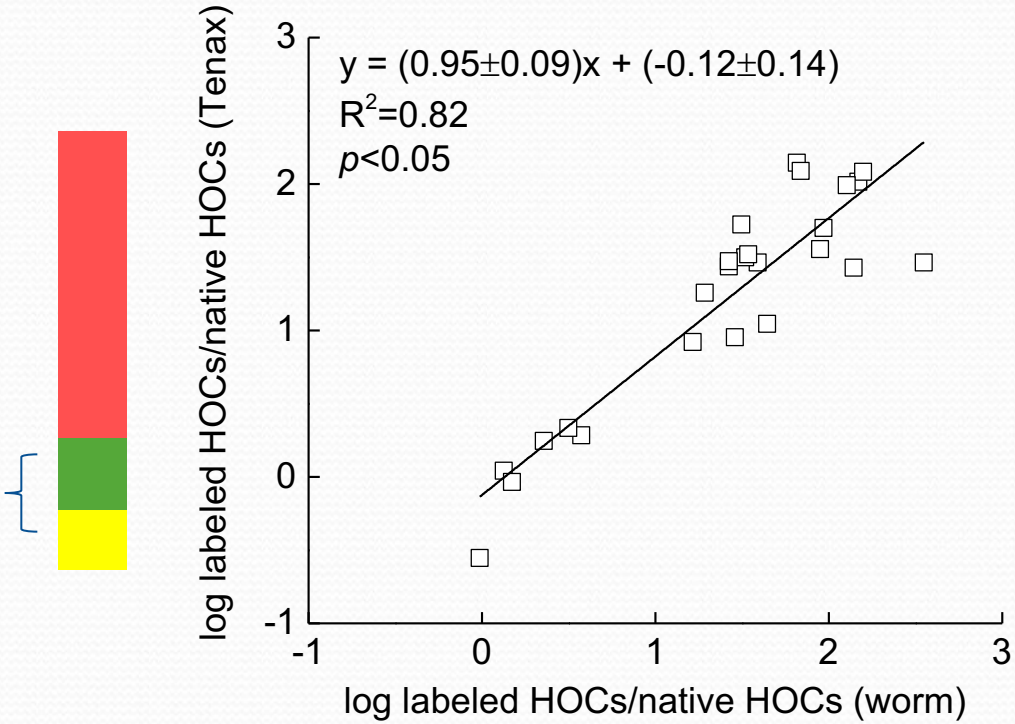
$$F_{24h} = \frac{C_{Tenax}}{C_{total}}$$

Tenax F_{24h} of native HOCs were consistently smaller than those for isotope labeled HOCs, suggesting reduced bioaccessibility due to aging



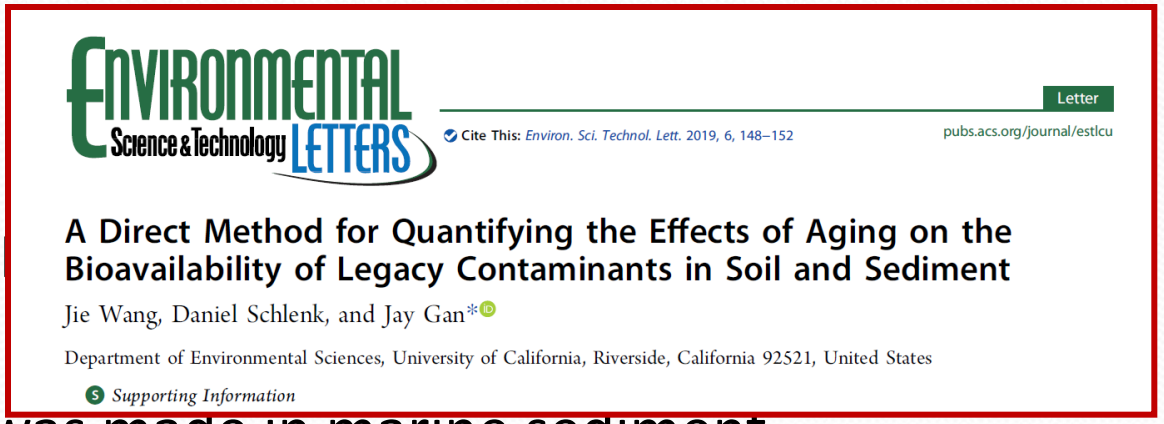
$$BSAF = \frac{C_{worm} / f_{lip}}{C_{total} / f_{oc}}$$

The BSAF values of native HOCs were consistently lower than those for isotope labeled HOCs, clearly indicating aging effect on bioavailability of POPs in environmental matrices



- Ratios of labeled HOCs to native HOCs accumulated in earthworm, against against with the ratios of Tenax F_{24h}.
- Highly significant linear correlation, with R^2 = 0.82, and slope close to 1.

Conclusions II



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Letter

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A Direct Method for Quantifying the Effects of Aging on the Bioavailability of Legacy Contaminants in Soil and Sediment

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Supporting Information

- Compared to freshly s
  aged residues was mu
- The same conclusion was made in marine sediment, freshwater sediment and soil, for different invertebrates
- The use of chemically based measurement closely predicted bioaccumulation
- The use of isotope labels is a direct and convincing technique to demonstrate the effect of aging on contaminant bioavailability.

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Review

Methods to assess bioavailability of hydrophobic organic contaminants: Principles, operations, and limitations

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