Using a risk-based approach to guide remedial goals: Oral relative bioavailability of PAHs at formerly used defense sites

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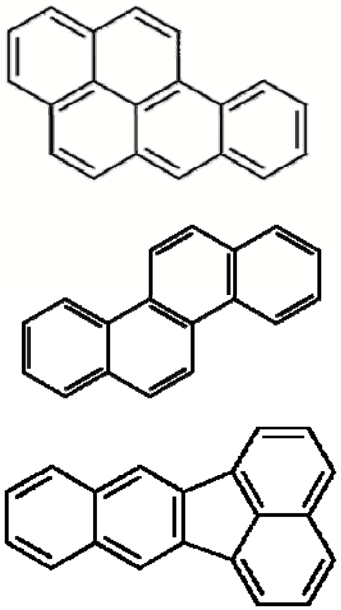
ISPRA
Sistema Nazionale per la Protezione dell'Ambiente
ANALISI DI RISCHIO DEI SITI CONTAMINATI
Opportunità e Prospettive a 10 anni dai "Criteri Metodologici"

Problem Statement

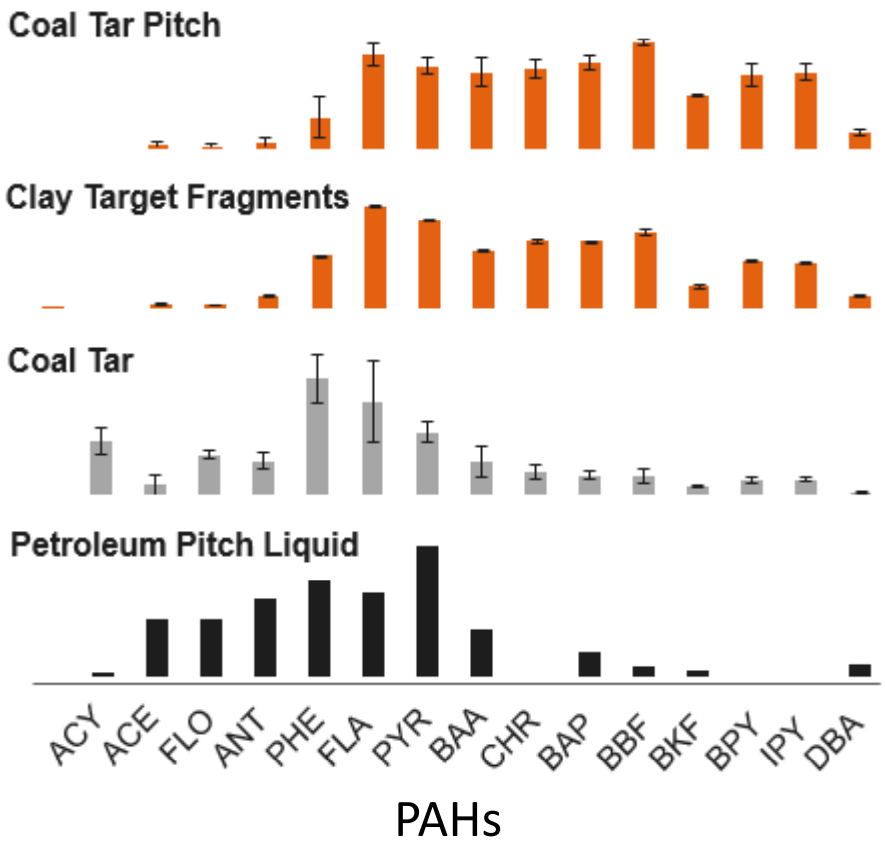
- The U.S. Department of Defense (DoD) is responsible for environmental restoration of formerly used defense sites
- Fragments of clay pigeon shooting targets accumulated in surface soil at shooting ranges
- Soils have concentrations of polycyclic aromatic hydrocarbons (PAHs) in excess of risk-based safety standards for human health
- DoD owns or operates over 3,000 of these sites

PAHs



Source of PAHs

- Formulation of historical clay pigeons:
  - 67% dolomitic limestone, 33% coal tar pitch as binding agent
- Coal tar pitch is the source of PAHs in targets
- High molecular weight PAHs are the chemicals of concern



Hypothesis and Rationale

- In the absence of site-specific information, risk assessments assume that substances are as bioavailable from soil as they were from the critical toxicology study from which their reference toxicity value was derived
- Dosing in the critical PAH toxicology study (Culp et al. 1998) used diets freshly spiked with PAHs dissolved in solvent
- Expect that oral bioavailability of PAHs will be less than achieved in Culp et al. (1998) based on physical-chemical properties of PAHs (i.e., log K_{ow}, V_P) and the PAH source (coal tar pitch is highly sorptive)

H: Oral bioavailability of PAHs for soils with clay pigeon target fragments is less than the oral bioavailability achieved for the critical PAH toxicology study

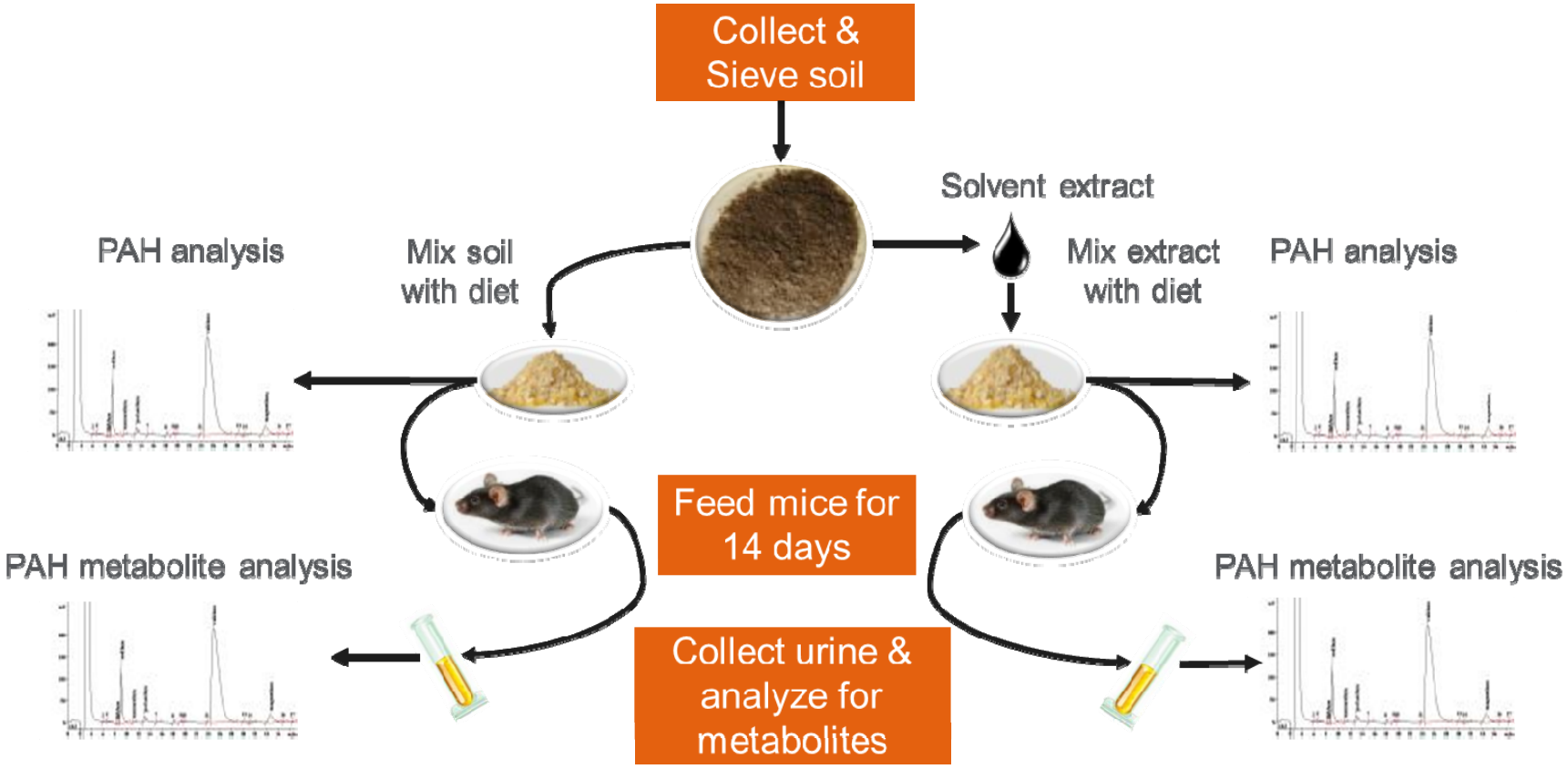
Sample Collection & Preparation

- 5 soils collected in bulk from a historical DoD shooting range
- Sieved (2 mm) to remove large fragments and debris
- Sieved to <250 µm and homogenized
- Data quality objective: Relative standard deviation of PAH concentrations <20% by GC-MS

Relative Bioavailability Testing

- Animal model: Female B6C3F1 mice (2 groups of 4 per group) (same as Culp et al. 1998)
- Diet preparation: 1) Soil-amended diet (5%), 2) Soil extract-amended diet ([PAHs] equivalent to soil-amended dose)
  - Concentration of PAHs in diets measured using GC-MS
- Exposure route, frequency, and duration: Oral, daily in diet for 14 days
- In-life measurements: Food consumption, body weight, and urine excretion
- Analysis: Urinary PAH metabolites by GC-MS (Woudneh et al. 2016)

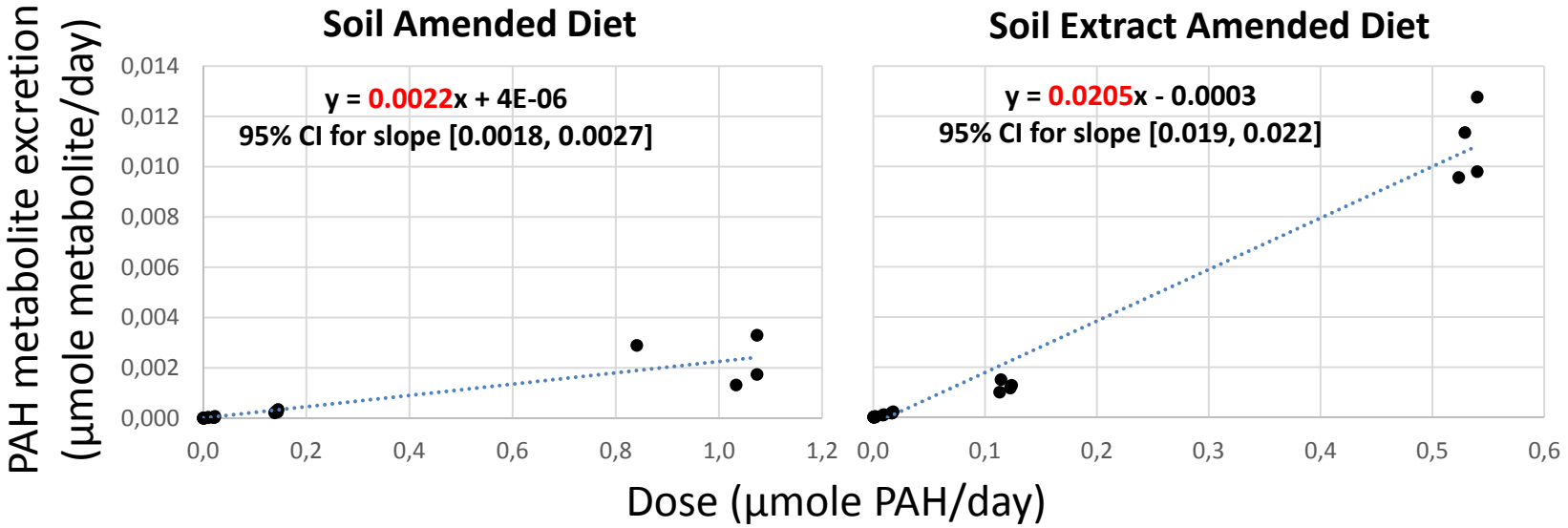
Testing Schematic



Relative Bioavailability Calculations

- Plot urinary excretion rate of PAH metabolites against PAH dose rates → Fraction of dose Eliminated in Urine (FUE)
- Perform regression analyses for (a) soil amended diets and (b) soil extract amended diets
- Calculate Relative Bioavailability Factor (RBAF) as ratio of two FUEs
- 95% confidence interval of mean RBAF using a Monte Carlo simulation approach

RBAF for Benzo(a)pyrene



FUE (Slopes) 0.0022 0.0205

$$\textbf{RBAF} = \frac{FUE_{Soil\ amended\ diet}}{FUE_{Soil\ extract\ amended\ diet}} = \frac{0.0022}{0.0205} = \textbf{0.11 (95\% CI: 0.09 to 0.13)}$$

Texas Remedial Goals

PAH	«Site A» (Commercial/Industrial)		«Site B» (Residential)	
	Default (mg/kg)	Site-Specific (mg/kg)	Default (mg/kg)	Site-Specific (mg/kg)
Benz(a)anthracene	170	880	41	170
Chrysene	---	---	4,100	15,000
Benzo(b)fluoranthene	170	980	41	190
Benzo(k)fluoranthene	1,700	12,000	420	2,100
Benzo(a)pyrene	17	43	4.1	19
Indeno(1,2,3-cd)pyrene	170	1,100	42	200
Dibenz(a,h)anthracene	17	120	4	17

Regulatory Status

- Texas Commission on Environmental Quality (TCEQ) is lead agency with input from USEPA
- TCEQ solicited peer-review from USEPA and an external bioavailability expert from the University of Florida
- Several White Papers were developed to provide scientific justification for the testing approach
- TCEQ reviewed and provided comments on all work plans
- **The methods and results were acceptable to reviewers**

*Note that a site-specific dermal absorption study was also performed and showed a reduction compared to the default assumption (e.g., default = 13% absorption, site-specific was <1% absorption).

Conclusions

- A reliable test was developed to evaluate oral bioavailability of PAHs for soil
- Results showed that PAHs in soils with historical clay pigeon shooting target fragments are much less bioavailable than default assumptions
- Site-specific bioavailability provides a more realistic evaluation and shows that PAH concentrations in soil can be ~10x times higher than defaults while achieving regulatory risk-based safety standards

The relative oral bioavailability test allowed for a more sustainable and cost-effective outcome for communities, the DoD, and the state of Texas

GRAZIE PER L'ATTENZIONE

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