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# USING A RISK-BASED APPROACH TO GUIDE REMEDIAL GOALS: ORAL BIOAVAILABILITY OF PAHS AT FORMERLY USED DEFENCE SITES

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# Problem Statement

- DoD is responsible for environmental restoration of formerly used defense sites (FUDS)
- Fragments of clay shooting targets accumulated in surface soil at trap and skeet shooting ranges
- Soils have concentrations of PAHs in excess of risk-based screening levels (by up to 5 orders or magnitude)
- DoD owns or operates over 3,000 of these sites


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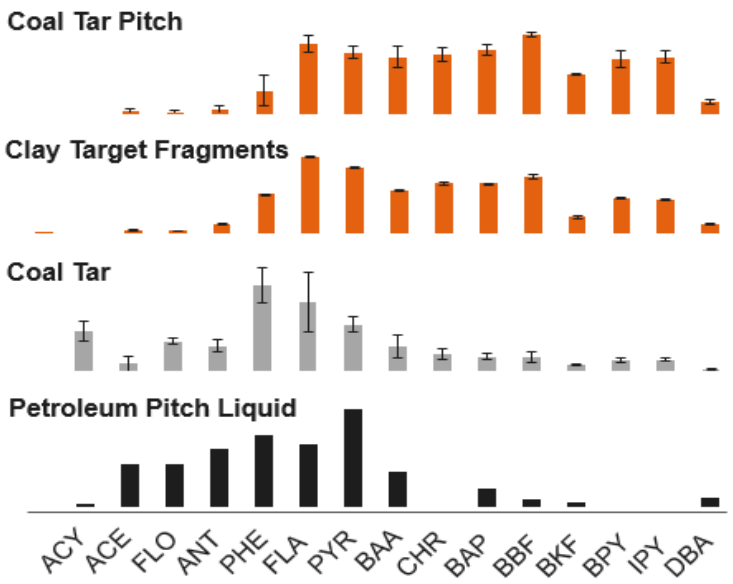
# Case Studies:

## Former Foster Air Force Base (Victoria, TX)

## Former Laredo Air Force Base (Laredo, TX)

# Source of PAHs

- 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





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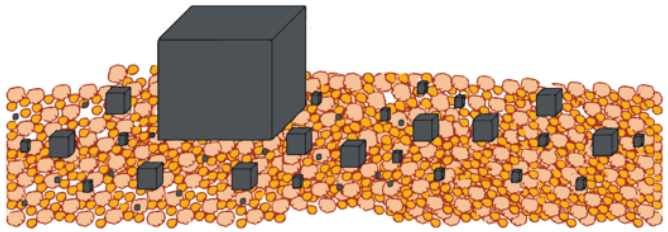
# Conceptual Model

- Mechanical weathering led to smaller target fragment pieces being incorporated into soil



| PAH | Concentration (mg/kg) of PAHs in Sieve Fractions (µm) | | | | |
|---|---|---|---|---|---|
| | >1000 & < 2000 | >250 & < 1000 | >150 & <250 | >50 & <150 | <50 |
| Fluoranthene | 380 | 170 | 90 | 84 | 71 |
| Pyrene | 400 | 160 | 53 | 85 | 68 |
| Benz(a)anthracene | 290 | 120 | 63 | 63 | 66 |
| Chrysene | 55 | 46 | 42 | 49 | 62 |
| Benzo(b)fluoranthene | 450 | 210 | 110 | 110 | 130 |
| Benzo(k)fluoranthene | 110 | 46 | 38 | 30 | 38 |
| Benzo(a)pyrene | 300 | 140 | 74 | 74 | 74 |
| Indeno(1,2,3-cd)pyrene | 260 | 120 | 68 | 67 | 75 |
| Dibenz(a,h)anthracene | 63 | 40 | 24 | 25 | 29 |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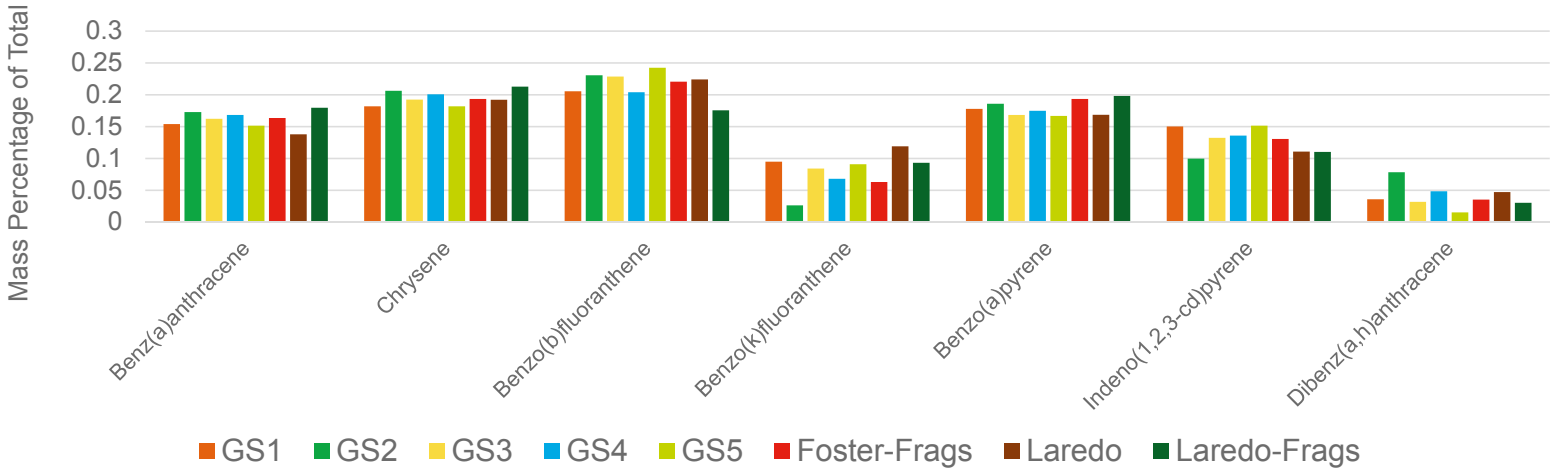
# PAH Profiles at Foster and Laredo

- Soil samples and target fragments collected from Foster and Laredo
- Homogenized and sieved samples to ≤250 µm for application to risk assessment → Analyze for PAHs





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# Default Exposure Scenarios

- Foster (Commercial/Industrial)
  - Incidental oral ingestion of soil (hand-to-mouth)
  - Direct dermal contact with soil
  - Inhalation of soil particles and/or volatiles
- Laredo (Residential)
  - Incidental oral ingestion of soil (hand to mouth)
  - Direct dermal contact with soil
  - Inhalation of soil particles and/or volatiles
  - Ingestion of home-grown vegetables


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# Hypothesis and Approach

Bioavailability of PAHs from soils with skeet target fragments is less than that observed in animal studies that used pure PAHs in solvents and which are the basis of reference toxicity values.

- *In vivo* Oral Bioavailability Study in Rodents
  - Default assumes bioavailability is equivalent to that achieved in reference toxicity study (rodent chow freshly spiked with dissolved BaP)


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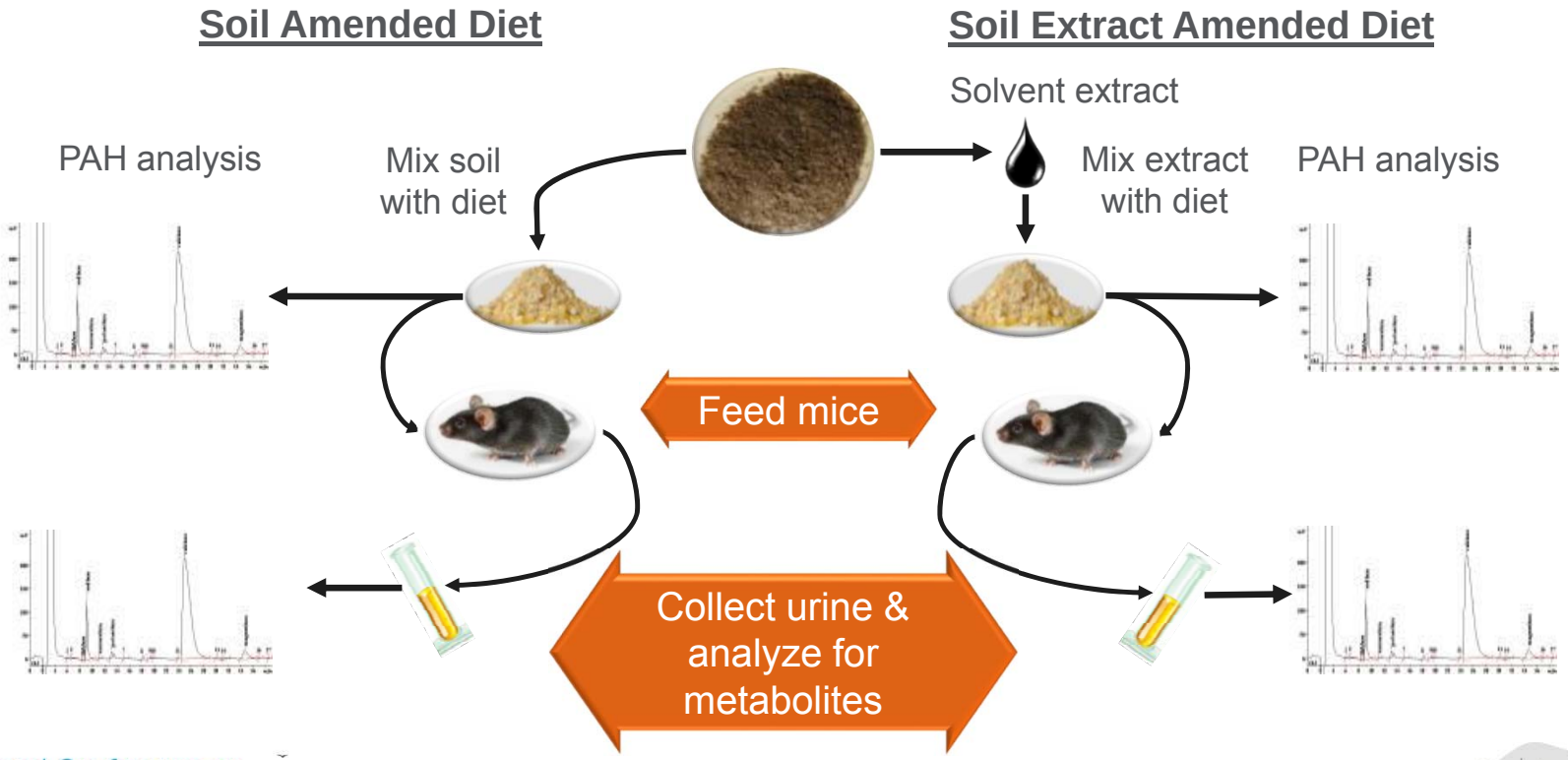
# *In Vivo* Oral Bioavailability Study

# Oral Bioavailability Overview





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# Sample Collection & Preparation

- Soils:
  - Bulk soil collected from locations across a skeet range
  - Sieved (2 mm) to remove large fragments and debris
  - Homogenize, sieved to <250 µm, rehomogenize
- Analyzed for PAHs by GC-MS
- RSD < 20% acceptability criterion


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# Experimental Design

- Diet preparation: 1) Soil-amended diet (5%), 2) Soil extract-amended diet ([PAHs] equivalent to soil dose)
- Animal model: Female B6C3F1 mice (2 groups of 4 per treatment)
- Exposure route, frequency, and duration: Oral, daily in diet for 14 days
- In-life measurements: Food consumption, body weight, and urine collection daily
- Analyzed for 3-OH-BaP, 9-OH-BaP, 3-OH-BaA, & 3-OH-chrysene by HRGC-HRMS


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# Calculations

- Calculate Fraction of Dose Eliminated in Urine (FUE) for Foster AFB and Laredo AFB samples
  - Perform regression analysis of data from both sites for (a) soil amended diets and (b) soil extract amended diets
- Calculate Relative Bioavailability Factor (RBAF) as ratio of two FUE slopes
- Determine 95% UCL for mean RBAF using a Monte Carlo approach


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ELSEVIER

Journal of Chromatography A

Volume 1454, 8 July 2016, Pages 93-100

# Quantitative determination of hydroxy polycylic aromatic hydrocarbons as a biomarker of exposure to carcinogenic polycyclic aromatic hydrocarbons

Million B. Woudneh [a], Jonathan P. Benskin [a], Richard Grace [a], M.Coreen Hamilton [a], Brian H. Magee [b], Glenn C. Hoeger [c], Norman D. Forsberg [b], John R. Cosgrove [a]



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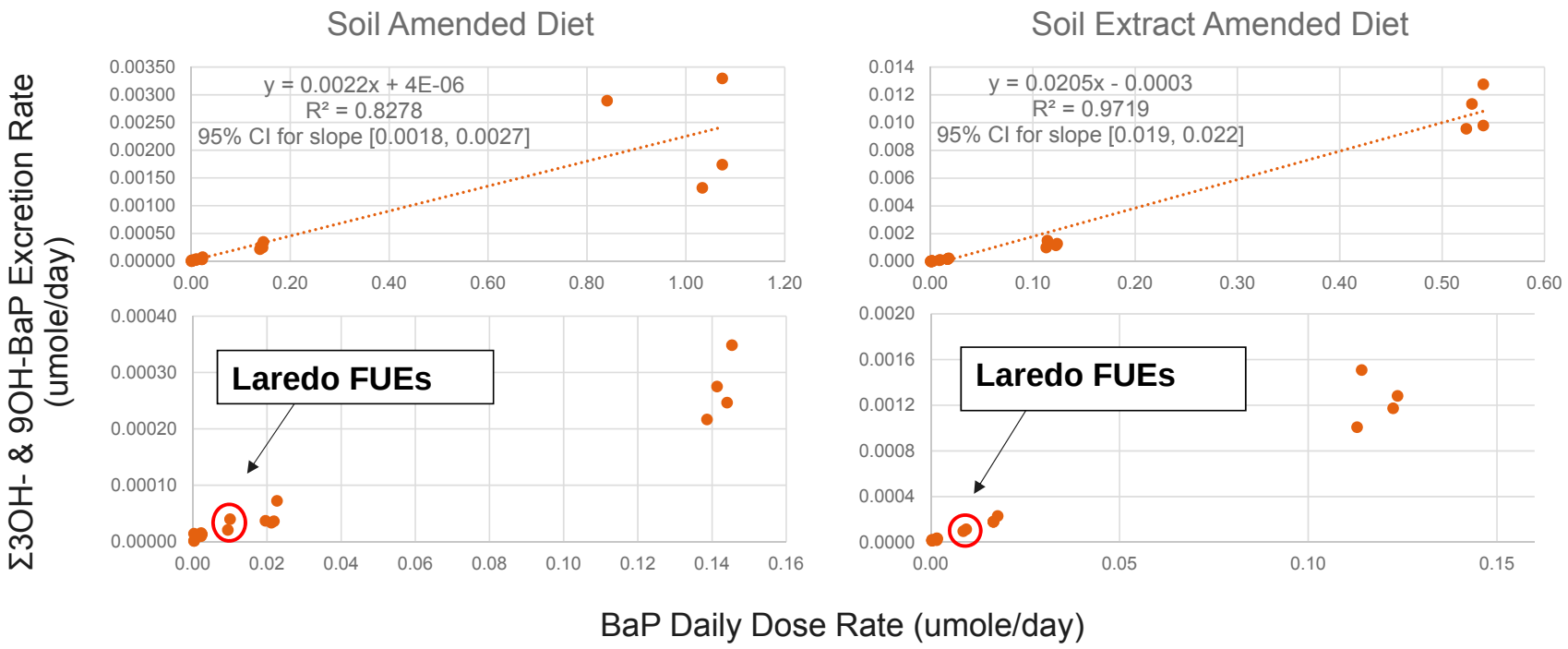
# Urinary Metabolite Excretion Rates





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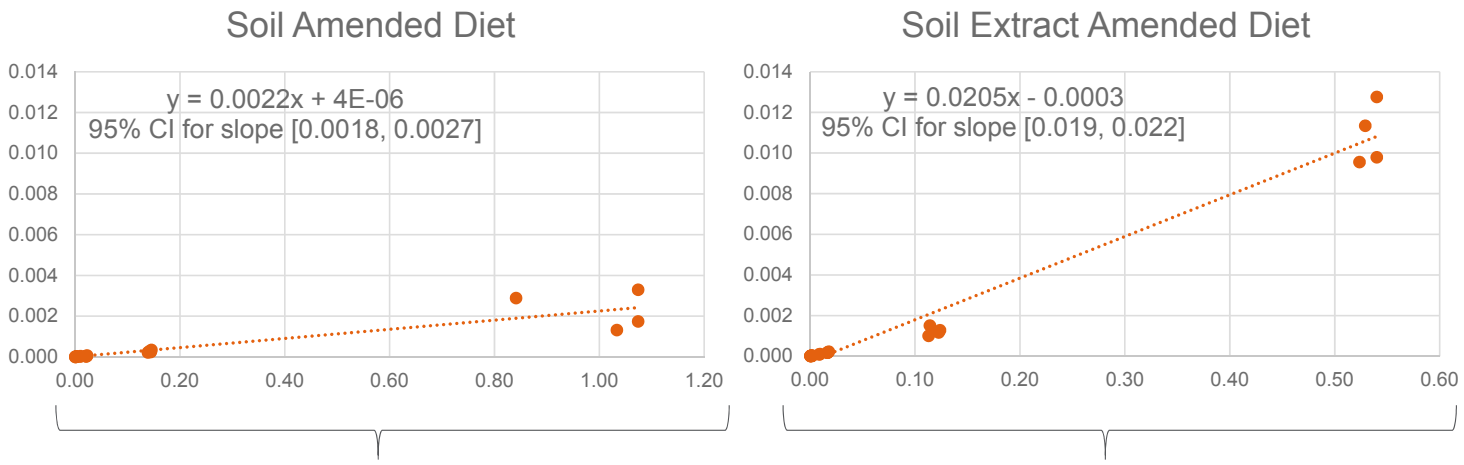
# Oral RBAF for BaP




**FUE Slopes** 0.0022 0.0205

**RBAF** $= \frac{FUE_{Soil\ amended\ diet}}{FUE_{Soil\ extract\ amended\ diet}} = \frac{0.0022}{0.0205} =$ 0.11 (95% UCL = 0.13) → **0.2**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# Oral RBAFs for PAHs

| PAH | RBAFs | Number Aromatic Rings | Molecular Weight (g/mole) | Log $K_{ow}$ | Water Solubility (µg/L) | Relative Potency |
|---|---|---|---|---|---|---|
| Benz(a)anthracene | 0.23 | 4 | 228 | 5.76 | 9.4 | 0.1 |
| Chrysene | 0.28 | 4 | 228 | 5.81 | 2.0 | 0.001 |
| Benzo(b)fluoranthene | 0.2 (BaP) | 5 | 252 | 5.78 | 1.5 | 0.1 |
| Benzo(k)fluoranthene | 0.2 (BaP) | 5 | 252 | 6.11 | 0.8 | 0.01 |
| Benzo(a)pyrene | 0.2 | 5 | 252 | 6.13 | 1.62 | 1 |
| Indeno(1,2,3-cd)pyrene | 0.2 (BaP) | 6 | 276 | 6.7 | 0.19 | 0.1 |
| Dibenz(a,h)anthracene | 0.2 (BaP) | 5 | 278 | 6.75 | 2.49 | 1 |

- Xia et al. (2016) showed that dermal absorption of PAHs primarily controlled by high sorption capacity of skeet targets, rather than soil characteristics


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# Effect on Remedial Investigations

# Texas Remedial Goals (PCLs)

| PAH | Foster (Commercial/Industrial) | | Laredo (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 |


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# Regulatory Engagement and Acceptance

- 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 the reviewers


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# Reduced Remedial Footprint

- Laredo (Properties):
  - Current-use scenario:
    - 6 properties exceeded defaults, none exceeded site-specific PCL values
  - Future-use scenario:
    - 12 properties exceeded defaults, 4 exceeded site-specific PCL values
- Foster (Grid cells):
  - 30 cells exceeded defaults, 12 exceeded site-specific PCL values
  - 95% UCL of site is within CERCLA acceptable risk range


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# Conclusions

- A reliable test was developed to evaluate oral bioavailability of PAHs
- Results showed that PAHs from soils with historical skeet targets are much less bioavailable than default assumptions
- Site-specific bioavailability provides a more realistic evaluation and shows that PAH concentrations in soil can be 3 to 7 times higher than defaults while achieving target regulatory safety standard
- Resulted in substantial reduction in remedial footprint at Laredo and Foster
- Provides avenue to more sustainable and cost effective outcome for communities, the Corps, and the state of Texas


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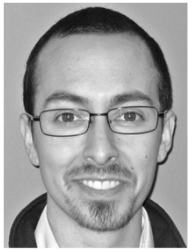
# Your Presenter(s)

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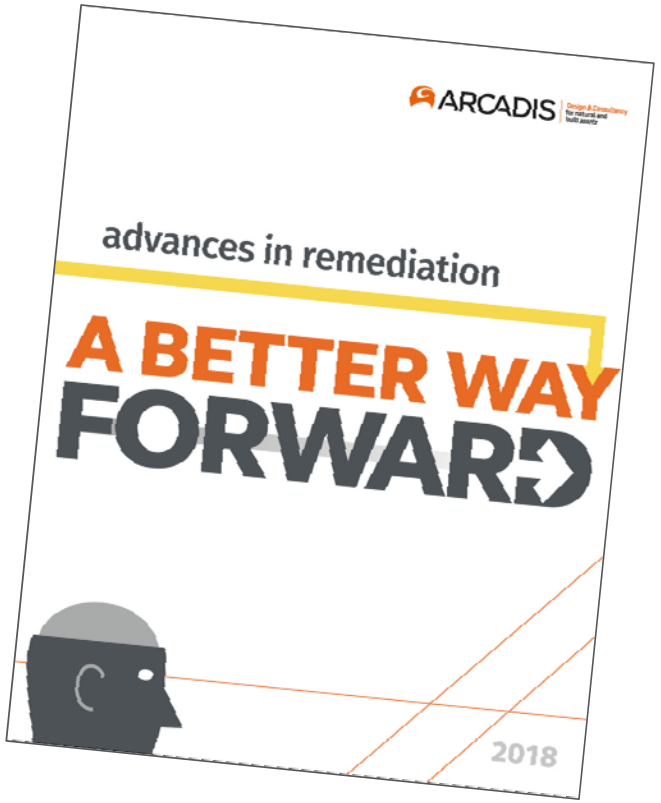



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