Laboratory Treatability Study on an Innovative Approach to Remediation of Groundwater Impacted with DNAPL Using Stabilized ClO₂/UV Radiation

by

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&

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Presentation Overview

- Problem Statement / Opportunity
- Beiseker Compressor Station – Soaking Pit
- Understanding DNAPL?
- Behavior of DNAPL in the Subsurface
- Objectives
- Treatability Lab Work Program
  - Physical Models....
- What is Next?
  - Field Pilot Remedial Design
- Questions?
Remediation of groundwater impacted by DNAPL is a significant challenge facing TransCanada and many industrial sectors in Alberta.

Why?

* Expensive
* Intrinsically Toxic
* Mutagenic / Carcinogenic
* Toxic Metabolites Formation
* Public / Regulatory Pressure
* Environmental / Financial Liabilities
Beiseker Compressor Station – Former Soaking Pit

- On-stream Date: 1970-10-26
- Two compressor buildings, a control building and a storage building.
- Original design had subsurface and building floor drains move fluids to a sump.
- These liquids were then pumped to a subsurface absorption pit or soaking pit.
- The former soaking pit measured approximately 21.0 m by 1.0 m, and consisted of a perforated pipe at a depth of 3.7 m to 4.3 m below grade, lined with crushed rock and covered with fill.
Beiseker Compressor Station – Former Soaking Pit

- Former Soaking Pit was removed in 1996.
- Soil and groundwater investigations have been ongoing since 2000.
- Analytical results from the collected DNAPL:
  - BTEX and PHC F1 to F4 concentrations are significant with a PHC F3 890,000 mg/l
  - PAHs, PCBs and Phenols are present at significant concentrations
  - The density analyses indicate that the product is about 15% denser than water and it has a kinematic viscosity of 69.57 cSt, which is greater than most lubricating oils
  - DNAPL is comprised of mainly phosphorus and sulphur, these results indicate the product is potentially a triaryl phosphate (TAP), Fyrquel fluid

Phase II Investigation
DNAPLs are denser than water allows them to migrate to substantial depths below the water table in both unconsolidated deposits and fractured bedrock.

The subsurface region containing residual and pooled DNAPL is referred to as the source zone.

As DNAPLs are only slightly soluble in water, DNAPL source zones can persist for many decades and, in some cases, even hundreds of years.
Residual DNAPL formation:
- Saturated: Unsaturated media
- Held in place by capillary and hydrophobic forces
- Adopt the shape of the aquitard
- Residual DNAPL retained is site-specific
  - A typical porous medium such as silt, sand and gravel is typically between 5 and 20 per cent of the pore space in the particular lenses and laminations invaded by the DNAPL.
The Opportunity

To develop innovative and cost-effective approach to risk manage groundwater contaminated with DNAPL
Components

Cost → Regulation → Public Relation → Innovation → Time
To ascertain:

* DNAPL constituents chemical degradation
* Dioxins and Furans formation
* DNAPL sorption / travel time....off site
* Physical model for more insights on tracer in the remediation design
* Reactions of fluorescein with ClO₂ at Room/GrH₂O temp, respectively
* Lab simulation for an infiltration gallery design

The above will inform the field pilot implementation!

* Ultimately, a field pilot!
Stock Concentrations: Phenol 20 µg/L; PCB 10 µg/L; PAH 20 µg/L; Gasoline 500 mg/L

<table>
<thead>
<tr>
<th>Treatment #</th>
<th>Phenol Addition (µL)</th>
<th>PCB Addition (µL)</th>
<th>PAH Addition (mL)</th>
<th>Gasoline Addition (µL)</th>
<th>UV Exposure</th>
<th>ClO₂ Addition</th>
<th>H₂SO₄ Addition*</th>
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<tbody>
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<td>24</td>
<td>1.2</td>
<td>48</td>
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<td>4</td>
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<td>48</td>
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<td>0</td>
<td>0</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
</tbody>
</table>

* 0.01 mL of 6M H₂SO₄ was added to treatment 4 in order to reduce the pH to 3.5
- Used clean groundwater from the site

- Initial Standard Analytes Concentration in the treatments:
  - Phenol: 20 µg/L
  - PAH: 20 µg/mL
  - Gasoline: 500 mg/mL
  - PCB: 20 µg/mL

- Blank, Control, Trts with or without ClO₂ and UV combination

All chemical analyses performed by Maxxam Analytics.
Batch Degradation Systems

\( \text{ClO}_2 / \text{UV} \)

**UV Lamp:**
- 254 nm
- 0.4 mW cm\(^{-2}\)
- 6 W / 100 V
- 11 inch

8 min Rxt time.
Stabilized ClO₂ Chemical?

* Strong Oxidant
* Water Soluble
* Oxidize over broad pH range
* Does not hydrolyze in H₂O
* No potential for toxic byproducts formation

\[
2 \text{ClO}_2 \text{(solid)} + \text{H}_2\text{O} \leftrightarrow \text{H}_2\text{O} + \text{ClO}_3^- + \text{ClO}_2^- \}
\]

Free radicals solution!
A very high oxidation capacity!

UV enhances quantum vibration in molecules & degradation by ClO₂!
Degradation Results

TRT 1: No degradation
TRT 2: 50-75% PCBs; 60-75% PAH; BETEX 50-80%; Phenol 74%
TRT 3: 97-100% PCBs; 76-93% PAH; BETEX 82-86%; Phenol 98%
TRT 4: 97-100% PCBs; 76-93% PAH; BETEX 82-86%; Phenol 98%

{\text{ClO}_2/UV + Soluble-DNAPL} \quad \rightarrow \quad \text{Simple-non toxic by-products + CO}_2 + \text{R-OH + Salts}

No formation of dioxins or furans as by-products was detected!
BATCH ISOTHERMS @ 6 °C

Contaminants Sorption

- Used clean subsoil substrate from the site 4.5-5 m
- Subsoil characterized for physical / chemical properties
- Spiked substrate with chemical standards / Equilibrated overnight
- Supernatant removed with glass syringe → Vial → Refrigerated

Textural Class = Clay
Sand =34%; Silt = 22%; Clay = 44%; pH = 8.1;
CEC = 19 cmole/kg; Ø = 60%; SAR = 0.66
BD= 1.23 gmL⁻¹; D₁₀ = 0.058; Cμ = 1.47;
CG = 0.0058

* Relative mobility / bioavailability / sorbing capacity evaluation

* Can model transport.....
## ISOTHERM EXPERIMENTAL DESIGN @ 6 °C

<table>
<thead>
<tr>
<th>Concentration of spiking solution (μg/mL)</th>
<th>Blank</th>
<th>Treatment 1</th>
<th>Treatment 2</th>
<th>Treatment 3</th>
<th>Treatment 4</th>
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<tr>
<td>Jar Weight (g)</td>
<td>183.3</td>
<td>181.13</td>
<td>185.19</td>
<td>185.19</td>
<td>188.25</td>
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<td>Sample weight (g)</td>
<td>79.92</td>
<td>79.65</td>
<td>80.15</td>
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<td>79.95</td>
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<td>Water Added (mL)</td>
<td>30</td>
<td>30</td>
<td>30</td>
<td>30</td>
<td>30</td>
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<tr>
<td>PAH (μL)</td>
<td>20</td>
<td>0</td>
<td>250</td>
<td>500</td>
<td>1000</td>
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<tr>
<td>PCB-S (μL)</td>
<td>10</td>
<td>0</td>
<td>50</td>
<td>100</td>
<td>200</td>
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<tr>
<td>SVPHEN-S (μL)</td>
<td>20</td>
<td>0</td>
<td>100</td>
<td>200</td>
<td>400</td>
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<tr>
<td>F24FIDE-S (μL)</td>
<td>70000</td>
<td>0</td>
<td>500</td>
<td>1000</td>
<td>2000</td>
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<tr>
<td>BTEXHSAB-S (μL)</td>
<td>500000</td>
<td>0</td>
<td>10</td>
<td>20</td>
<td>40</td>
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<tr>
<td>Total Volume Spiked (mL)</td>
<td></td>
<td></td>
<td>910</td>
<td>1820</td>
<td>3640</td>
</tr>
<tr>
<td>Total volume added to soil samples and total volume of water reference samples (mL)</td>
<td></td>
<td></td>
<td>30.91</td>
<td>31.82</td>
<td>33.64</td>
</tr>
</tbody>
</table>
Freundlich: \[ X/M = K \text{Ceq}^{1/n} \]

\[ \Rightarrow \log \frac{X}{M} = \frac{1}{n} \log \text{Ceq} + \log [Kd] \]
\[ \Rightarrow K_{oc} = Kd f_{oc} \]

----------------------------------------------

Langmuir: \[ X/M = \frac{(ab)(\text{Ceq})}{1 + b\text{Ceq}} \]

\[ \Rightarrow \frac{\text{Ceq}}{X/M} = \frac{1}{ab} + \frac{\text{Ceq}}{a} \]
\[ V = \frac{K \, (\text{dh/dl})}{(7.48 \, \emptyset) \, R_f} \]

Freundlich \[ \Rightarrow \quad R_f = 1 + \left\{ \frac{[Pb/\Phi]}{Kd} \right\} \]

Langmuir \[ \Rightarrow \quad R_f = 1 + \left\{ \frac{[Pb/\Phi]}{} \right\} \left\{ \frac{a \, b}{(1 + a \, C_{eq})^2} \right\} \]
### Freundlich & Langmuir Linear Regression Model Coefficients

<table>
<thead>
<tr>
<th>Analytes</th>
<th>Linear Regression</th>
<th>R2</th>
<th>Langmuir</th>
<th>R2</th>
<th>a (mg/Kg)</th>
<th>b (mL/g)</th>
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<tbody>
<tr>
<td></td>
<td>y = 38.969x + 9.8344</td>
<td>2.0759</td>
<td>0.9688</td>
<td>y = -0.0057x + 49.433</td>
<td>0.003</td>
<td>-175.4385965</td>
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<tr>
<td>TOLUENE</td>
<td>y = 113.47x - 13.293</td>
<td>2.2267</td>
<td>0.9736</td>
<td>y = -0.005x + 50.378</td>
<td>0.003</td>
<td>-200</td>
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<tr>
<td>ETHYLBENZENE</td>
<td>y = 473.54x - 46.407</td>
<td>2.6529</td>
<td>0.9786</td>
<td>y = -0.0048x + 51</td>
<td>0.002</td>
<td>-208.3333333</td>
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<tr>
<td>m &amp; p-XYLENE</td>
<td>y = 115.86x - 40.364</td>
<td>2.0786</td>
<td>0.9729</td>
<td>y = -0.0047x + 50.095</td>
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<td>-212.7659574</td>
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<tr>
<td>o-XYLENE</td>
<td>y = 312.89 - 34.513</td>
<td>2.471</td>
<td>0.9775</td>
<td>y = -0.0048x + 50.824</td>
<td>0.002</td>
<td>-208.3333333</td>
</tr>
<tr>
<td>F1 (C6-C10)</td>
<td>y = 3.2528x + 90.15</td>
<td>2.0252</td>
<td>0.8325</td>
<td>y = -0.0087x + 184.19</td>
<td>0.003</td>
<td>-114.9425287</td>
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<tr>
<td>PAH-ACENAPHTENE</td>
<td>y = 4.3599x - 0.111</td>
<td>0.892</td>
<td>0.8784</td>
<td>y = 0.0113x + 0.0184</td>
<td>0.043</td>
<td>88.49557522</td>
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<tr>
<td>PAH-ACENAPHTYLENE</td>
<td>y = 4.8059x - 0.1076</td>
<td>1.0766</td>
<td>0.8983</td>
<td>y = 0.0098x + 0.0202</td>
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<td>102.0408163</td>
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<tr>
<td>PAH-ACRIDINE</td>
<td>y = 4.6739x - 0.1044</td>
<td>1.037</td>
<td>0.8949</td>
<td>y = 0.0092x + 0.0204</td>
<td>0.026</td>
<td>108.6956522</td>
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<tr>
<td>PAH-ANTHRACENE</td>
<td>y = 4.7797x - 0.1056</td>
<td>1.0172</td>
<td>0.8929</td>
<td>y = 0.0092x + 0.0207</td>
<td>0.024</td>
<td>108.6956522</td>
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<tr>
<td>PAH-BENZO(a)ANTHRACENE</td>
<td>y = 4.6154x - 0.1021</td>
<td>0.9978</td>
<td>0.8913</td>
<td>y = 0.0085x + 0.0206</td>
<td>0.019</td>
<td>117.6470588</td>
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<tr>
<td>PAH-BENZO(k)FLUORANTHACENE</td>
<td>y = 4.904x - 0.1025</td>
<td>1.0114</td>
<td>0.9072</td>
<td>y = 0.0077x + 0.0222</td>
<td>0.010</td>
<td>147.0588235</td>
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<tr>
<td>PAH-BENZO(C)PHENANTHRENE</td>
<td>y = 4.6398x - 0.097</td>
<td>0.9588</td>
<td>0.8999</td>
<td>y = 0.0068x + 0.0222</td>
<td>0.010</td>
<td>147.0588235</td>
</tr>
<tr>
<td>PAH-BENZO(e)PYRENE</td>
<td>y = 5.3072x - 0.1039</td>
<td>0.9917</td>
<td>0.9053</td>
<td>y = 0.006x + 0.0248</td>
<td>0.008</td>
<td>166.6666667</td>
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<tr>
<td>PAH-CHRYSENE</td>
<td>y = 6.5006x - 0.1054</td>
<td>1.0607</td>
<td>0.8843</td>
<td>y = 0.0125x + 0.017</td>
<td>0.058</td>
<td>80</td>
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<tr>
<td>PAH-FLUORANTHENE</td>
<td>y = 4.6268x - 0.1063</td>
<td>1.0163</td>
<td>0.8853</td>
<td>y = 0.0205x + 0.019</td>
<td>0.034</td>
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<tr>
<td>PAH-FLUORENE</td>
<td>y = 4.3412x - 0.1055</td>
<td>1.0659</td>
<td>0.8775</td>
<td>y = 0.0137x + 0.0154</td>
<td>0.078</td>
<td>72.99270073</td>
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<tr>
<td>PAH-2-METHYLNAPHTALENE</td>
<td>y = 5.8116x - 0.1004</td>
<td>0.9971</td>
<td>0.9311</td>
<td>y = 0.0066x + 0.0342</td>
<td>7E-05</td>
<td>166.6666667</td>
</tr>
<tr>
<td>PAH-NAPHTALENE</td>
<td>y = 6.066x - 0.0985</td>
<td>0.9852</td>
<td>0.9372</td>
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<tr>
<td>PAH-PHENANTHRENE</td>
<td>y = 4.204x - 0.1128</td>
<td>1.2091</td>
<td>0.8633</td>
<td>y = 0.0189x + 0.0111</td>
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<td>PAH-PERYLENE</td>
<td>y = 4.482x - 0.1133</td>
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<td>0.8694</td>
<td>y = 0.0153x + 0.0146</td>
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<td>PAH-PYRENE</td>
<td>y = 4.347x - 0.1091</td>
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<td>0.8685</td>
<td>y = 0.0152x + 0.0144</td>
<td>0.103</td>
<td>65.78947368</td>
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<tr>
<td>PHE-CRESOLS</td>
<td>y = 13.054x - 0.067</td>
<td>3.0488</td>
<td>0.8169</td>
<td>y = 0.0079x + 0.0113</td>
<td>0.021</td>
<td>126.5822785</td>
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<tr>
<td>PHE-2,3,5,6-TETRACHLOROPHENOL</td>
<td>y = 9.2898x - 0.0408</td>
<td>2.2951</td>
<td>0.8483</td>
<td>y = -0.0043x + 0.0121</td>
<td>0.002</td>
<td>-232.5581395</td>
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<td>PHE-2,3,4,6-TETRACHLOROPHENOL</td>
<td>y = 8.5953x - 0.038</td>
<td>2.5427</td>
<td>0.8781</td>
<td>y = -9E-05 + 0.0097</td>
<td>2E-06</td>
<td>11111.11111</td>
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<td>PHE-2,4-DICHLOROPHENOL</td>
<td>y = 7.5085x - 0.0395</td>
<td>2.3928</td>
<td>0.7629</td>
<td>y = 0.0031x + 0.0081</td>
<td>0.001</td>
<td>322.5806452</td>
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<td>PHE-2,6-DICHLOROPHENOL</td>
<td>y = 2.0592x + 0.1705</td>
<td>1.5799</td>
<td>0.0102</td>
<td>y = 0.7926x - 0.0618</td>
<td>0.081</td>
<td>126.1670452</td>
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<td>PHE-2,CHLOROPHENOL</td>
<td>y = 11.131x - 0.049</td>
<td>2.3212</td>
<td>0.8412</td>
<td>y = -0.005x + 0.0149</td>
<td>0.002</td>
<td>-200</td>
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<tr>
<td>PHE-3 &amp; 4-METHYLPHENOL</td>
<td>y = 13.054x - 0.067</td>
<td>3.0488</td>
<td>0.8169</td>
<td>y = 0.0079x + 0.0113</td>
<td>0.0216</td>
<td>126.5822785</td>
</tr>
</tbody>
</table>
Both Freundlich and Langmuir isotherm equations evaluated. Better represented by Freundlich model:

\[ R^2 \text{ ranged from } 0.76 \text{ to } 0.98 \]
\[ K_d \text{ ranged from } 0.89 \text{ to } 3.05 \]

GLMs will be evaluated also!
Both Freundlich and Langmuir isotherm equations evaluated. 
Better represented by Freundlich model:

\[ R^2 \text{ ranged from 0.76 to 0.98 } \]
\[ K_d \text{ ranged from 0.89 to 3.05 } \]

GLMs will be evaluated also!
Equation for Predicting Advection Transport

\[ V_s = \frac{Q}{A \Theta} R \quad \text{"} K \frac{dh}{dL} / \Theta R \]

\[ R = \text{Retardation factor} \]

Freundlich, \( R = 1 + \left\{ \frac{pb}{\Theta} \right\} kd \)

Langmuir, \( R = 1 + \left[ \frac{pb}{\Theta} \right] \frac{ab}{(1 + a C_{eq})^2} \)

\[ \text{Travel Time} = \frac{L}{V_s} \]
Pertinent Information from Phase II investigation:

- Ground H2O elevations
- Flow direction
- Isopleth of equipotential lines
- Slug test method:
  * $K$ values ranged from $1.87 \times 10^{-7}$ to $1.30 \times 10^{-5}$ m/s
  * $K$ & $\bar{K}$ as per Falling-Head Method; $\bar{K}$; BD
<table>
<thead>
<tr>
<th>Contaminant Isotherm</th>
<th>Freundlich KD</th>
<th>BH-20</th>
<th>BH-21</th>
<th>BH-23</th>
<th>BH-20</th>
<th>BH-21</th>
<th>BH-23</th>
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<td>BENZENE</td>
<td>2.0759</td>
<td>17245.33</td>
<td>2328.68</td>
<td>288.56</td>
<td>11847.40</td>
<td>1744.91</td>
<td>288.56</td>
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<td>TOLUENE</td>
<td>2.2267</td>
<td>18254.92</td>
<td>2469.42</td>
<td>306.78</td>
<td>12159.91</td>
<td>1850.36</td>
<td>306.78</td>
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<td>ETHYLBENZENE</td>
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<td>21106.28</td>
<td>2867.17</td>
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<td>14060.57</td>
<td>2148.40</td>
<td>358.27</td>
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<td>m &amp; p- XYLENE</td>
<td>2.0786</td>
<td>17263.41</td>
<td>2331.20</td>
<td>288.89</td>
<td>11494.44</td>
<td>1746.80</td>
<td>288.89</td>
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<tr>
<td>o-XYLENE</td>
<td>2.471</td>
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<td>2697.41</td>
<td>336.30</td>
<td>13249.38</td>
<td>2021.20</td>
<td>336.30</td>
</tr>
<tr>
<td>F1 (CG-C10)</td>
<td>2.0252</td>
<td>16905.90</td>
<td>2281.37</td>
<td>282.44</td>
<td>11261.30</td>
<td>1709.46</td>
<td>282.44</td>
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<tr>
<td>PAH-ACENAPHTENE</td>
<td>0.892</td>
<td>9319.27</td>
<td>1223.81</td>
<td>145.53</td>
<td>6207.72</td>
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<td>PAH-ACENAPHTYLENE</td>
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<td>7030.96</td>
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<td>167.83</td>
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<tr>
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Infiltration Gallery Design
**PROPOSED INFILTRATION GALLERY SYSTEM (NTS)**

- **Infiltration Gallery**
  - **MW 01A**
  - Depth = 6m
  - Flow Rate = 17 IGN

- **Infiltration Gallery**
  - **MW**
  - Depth = 6m
  - Flow Rate = 12 IGN

- **R. Well**

- **Infiltration Gallery**
  - **MW 01A**
  - Depth = 4m
  - Flow Rate = 8 IGN

- **Reintroduction Wells**
  - 12" Diameter
  - Down gradient

**ClO₂ Treatment Method**

- Aboveground NAPL degradation
  - ClO₂ and UV exposure
- Rejection of treated groundwater with 10ppm ClO₂
- Radius of influence overlaps by 4" (shown below)
**Infiltration Gallery Profile (NTS)**

- **Air Compressor**
- **Flow Rate Control Valve**
- \( \frac{3}{4} \) "PVC Pipe"
- **Injection Ports**
  - \( \frac{1}{2} \) "PVC piping (perforated)"
- **Capped Bottom**

---

**Infiltration Gallery Notes**
- Dedicated pump and treatment systems will be used for each monitoring well
- Treatment systems will be sheltered
- Reinjection of treated ground water will be up gradient
- Field monitoring of residual ClO₂ in the ground water will be performed
Fluorescein Tracer Applications

- Slightly soluble in water
- Visually detected
- Fluorophore
- Exhibit a yellow color/can appear red
- Non-toxic
- Inert
- Analysed with a fluorometer / turbidimeter
Arrival time reflects advective transport through the physical model.
Batch aboveground treatment System

↓

Reinjection of treated \( \text{GrH}_2\text{O} + \text{ClO}_2 \)

↓

Network Infiltration Gallery

Ongoing Monitoring........
Conclusions

- ClO$_2$/UV effective treatment system for dissolved DNAPL plume
- No detectable dioxins or furans as by-products of the chemical reactions
- Travel time ranged from 145 to 80,817 years
- Public health risk & financial liabilities can be cost effectively / confidently managed
Thank You!

Questions?