

# Welcome to the Dense Non-Aqueous Phase Liquids (DNAPLs) Site Characterisation: Tools and Techniques Webinar

Presented by:

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Geosyntec<sup>®</sup>  
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Dr Mike Rivett  
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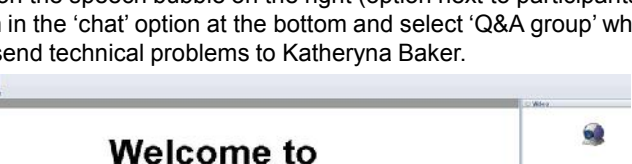
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(asking questions) 



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# DNAPL Site Characterisation: Tools and Techniques

## 22 September 2011

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## Instructors

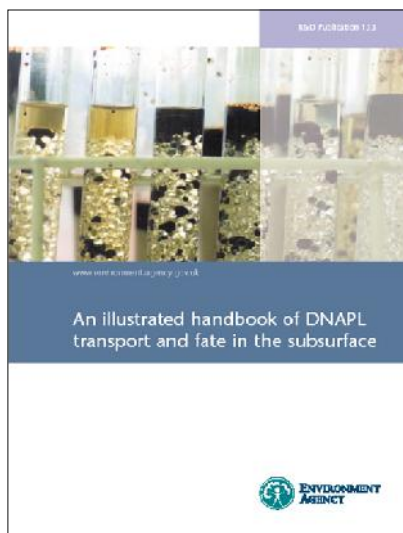
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## Background reading



<http://publications.environment-agency.gov.uk/pdf/SCHO0604BHIT-e-e.pdf>




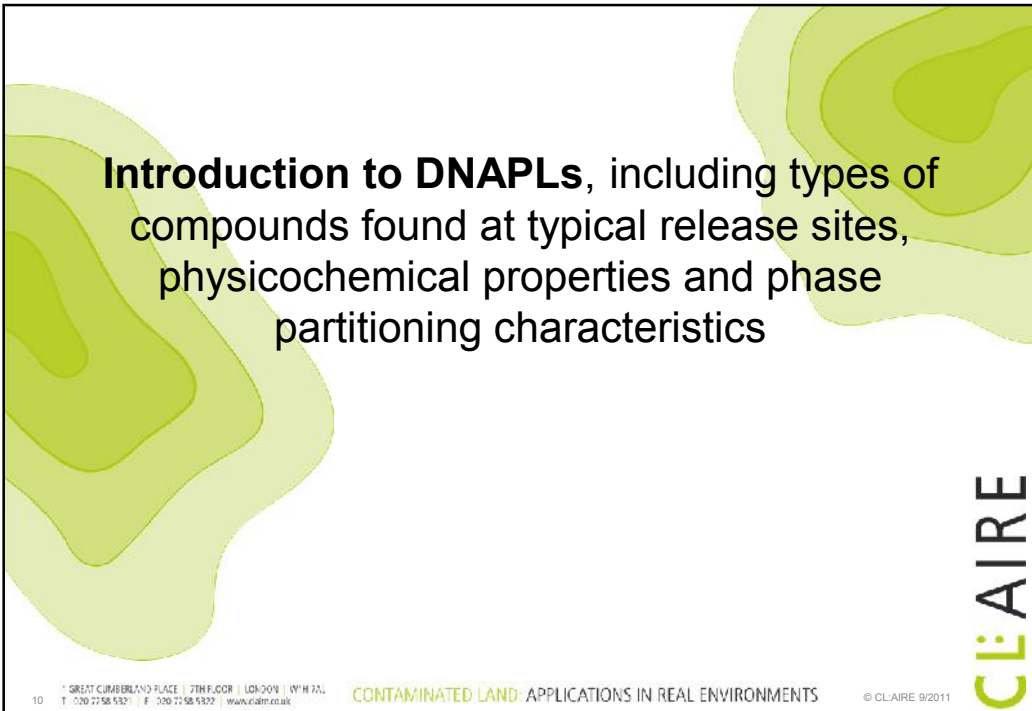
## Overview

- 1. Introduction to DNAPLs**, including types of compounds found at typical release sites, physicochemical properties and phase partitioning characteristics
- 2. Developing DNAPL conceptual site models** for soil and bedrock
- 3. Characterising DNAPL source zones**, including direct and indirect tools, emerging investigation technologies and selection of which parameters to measure at DNAPL sites
- 4. Calculating DNAPL mass estimates** from typical site investigation data. Understanding and communicating uncertainty in contaminant mass distribution to stakeholders
- 5. Summary of available guidance documents** along with key academic literature

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## Introduction to DNAPLs, including types of compounds found at typical release sites, physicochemical properties and phase partitioning characteristics

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# DNAPLs

## Dense non-aqueous phase liquids



DNAPL class	Main physical properties	Example	Principal uses
Chlorinated hydrocarbons	Density: 1100 to 1630 kg m <sup>-3</sup> Viscosity: around 1 cP Solubility: low Sorption to aquifer minerals: low	Tetrachloroethene Trichloroethene	Dry-cleaning fluid Degreaser
Coal tar or creosote	Density: 1010 to 1050 kg m <sup>-3</sup> Viscosity: 10 to 100 cP Solubility: low Sorption to aquifer minerals: Variable	Creosote Coal tar	Wood preservative Waste product from coal gasification
Polychlorinated biphenyls (PCBs)	Density: 1180 to 1420 kg m <sup>-3</sup> Viscosity: 5 to 65 cP (Aroclor oils) Solubility: low Sorption to aquifer minerals: high	Aroclor 1254	Insulating oil
Pesticides	Density: 1000 to 2000 kg m <sup>-3</sup> Viscosity: highly variable Solubility: variable Sorption to aquifer minerals: variable	Chlordane	Insecticide



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# 1950s Love Canal (NY)

## Chlorinated pesticides :: PCBs :: Benzene

- Miscarriages
- Birth defects
- Nervous disorders
- Cancers



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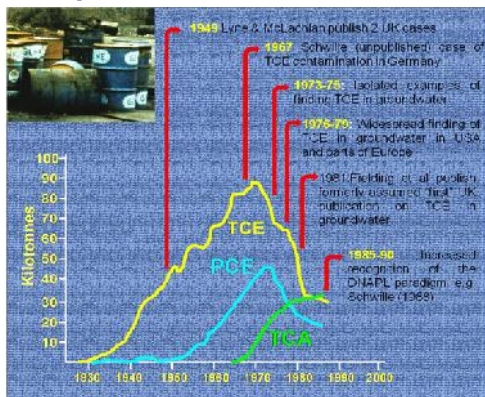
<http://library.buffalo.edu/libraries/specialcollections/lovecanal/gallery.html#thumb>

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## History matters

### UK production estimate



- 'Safe' disposal often not practiced/regulated until 1980s
- Research community not aware of DNAPLs until early 1980s (LNAPLs in 1950s)
- Currently 100,000s of sites across North America, Europe, and Asia

Rivett M.O., et al., 2006. Lyne and McLachlan (1949): Influence of the first publication on groundwater contamination by trichloroethene. *Environmental Forensics* 7(4), 313-323.

## Industries With High Probability of Historical DNAPL Release

- Electronics manufacturing
- Solvent production
- Dry cleaning
- Timber treatment
- Steel industry coking
- Aeroplane maintenance
- Engine/Vehicle manufacturing
- Pesticide/herbicide manufacturing
- Military bases & rocket fuel production
- Coal gasification (town gas production)
- Transformer oil production/reprocessing



(USEPA, 1992)

## Chlorinated Solvents

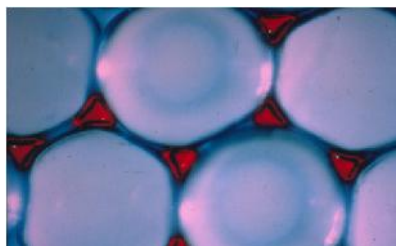
- Some of the most frequently detected chemicals in groundwater
- Typical uses: Dry cleaning, metal degreasing, pharmaceutical production, pesticide formulation, chemical manufacturing, electronics manufacturing
- Chlorinated ethenes
  - Perchloroethene (PCE)
  - Trichloroethene (TCE)
  - 1,1,1-Trichloroethane (TCA)



## DNAPL mass distribution controls

### Properties

- Density
- Viscosity
- Interfacial tension
- Contact angle
- Capillary pressure



From Schwille (1988)

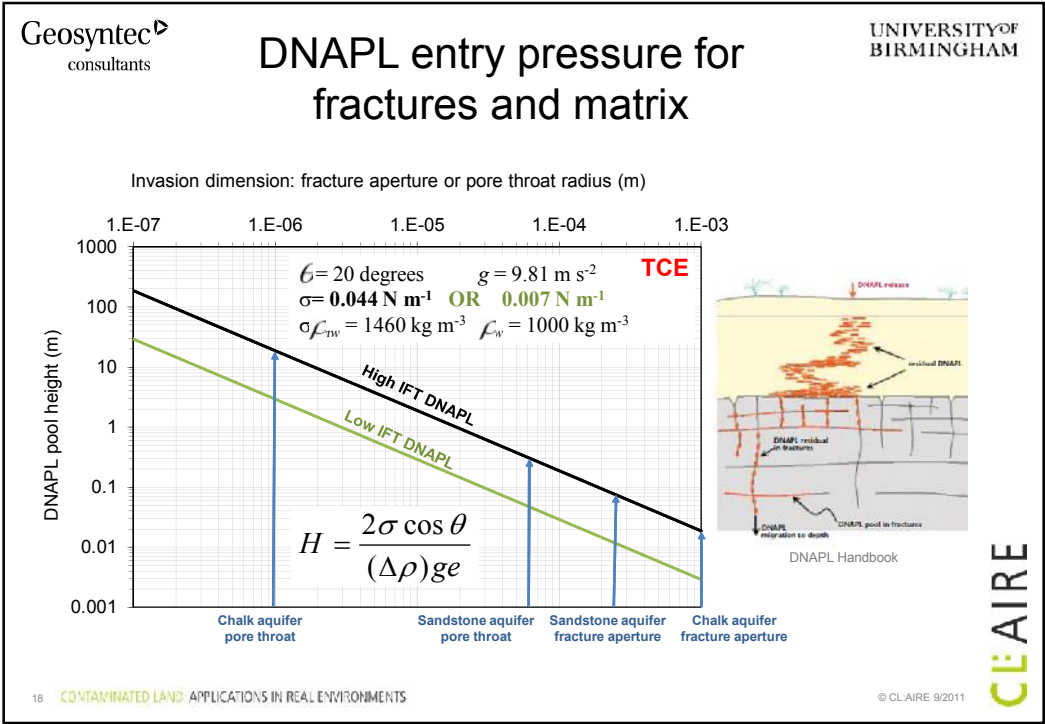
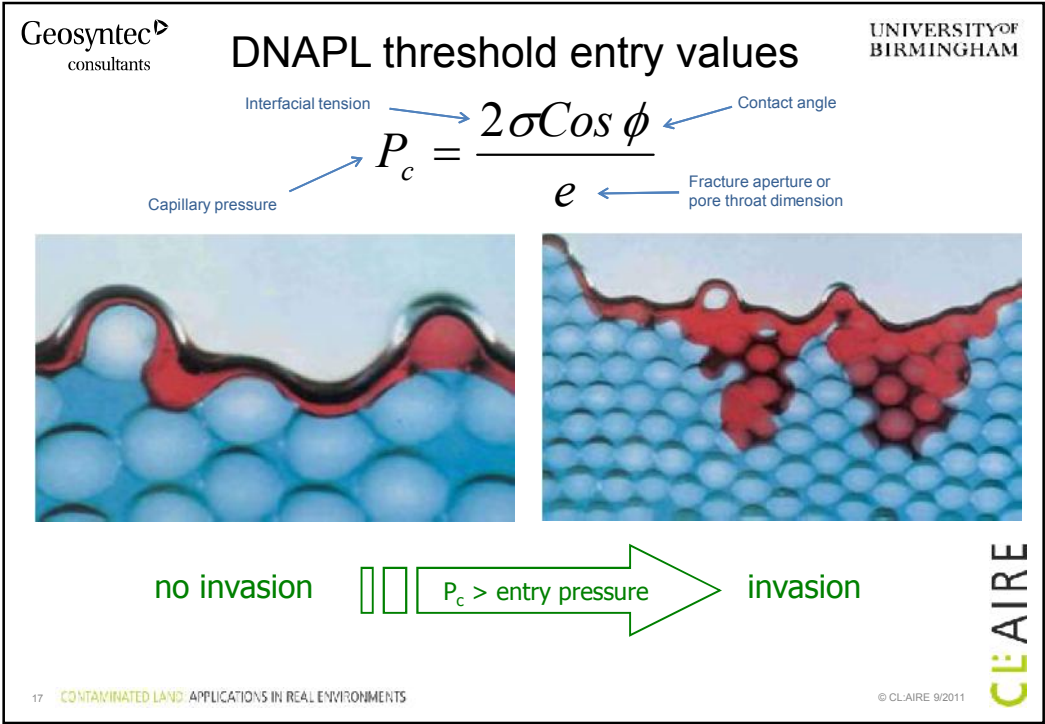
### Mass transfer processes

- Dissolution
- Vapourisation



Courtesy of Jason Gerhard





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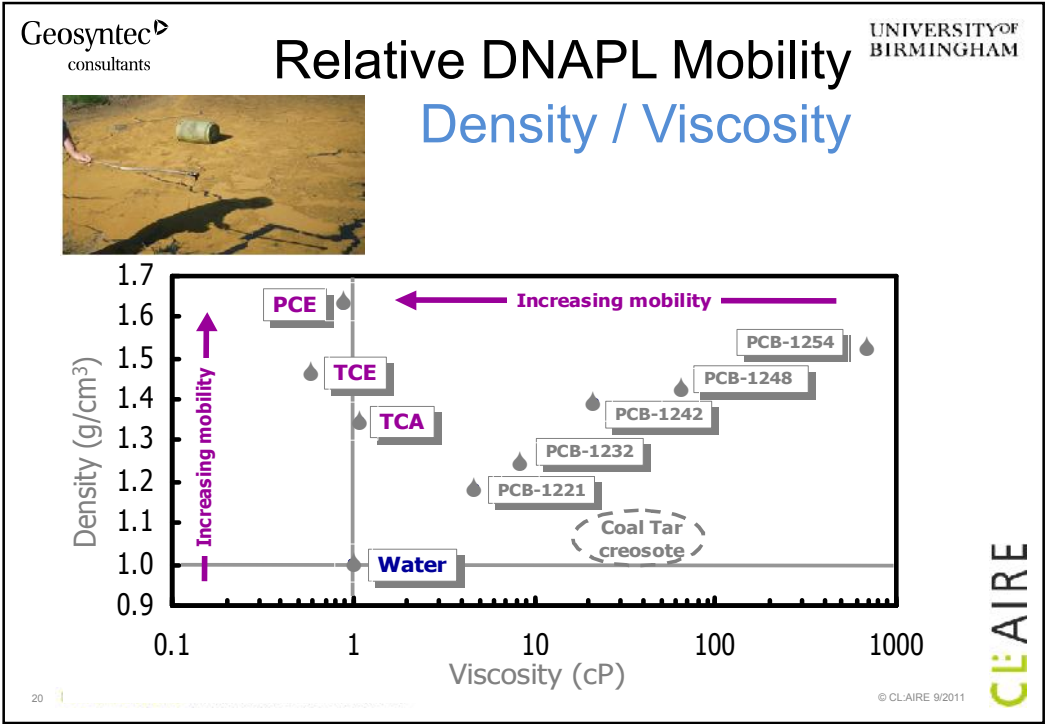
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Relative DNAPL Mobility  
Density / Viscosity

Chlorinated Organic Solvent	Density (g/cm <sup>3</sup> )	Viscosity (cP)	Mobility (s/cm <sup>2</sup> )
1,2 Dichloroethane	1.25	0.84	1.47
1,1,1-Trichloroethane	1.35	0.84	1.59
Carbon tetrachloride	1.59	0.97	1.64
Chloroform	1.49	0.56	2.66
Tetrachloroethene (PCE)	1.63	0.90	1.81
Trichloroethene (TCE)	1.46	0.57	2.56

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Permeability control


- mm / cm control on DNAPL migration
- Heterogeneous source zone architecture



DNAPL PCE

in Borden sands / aquitard





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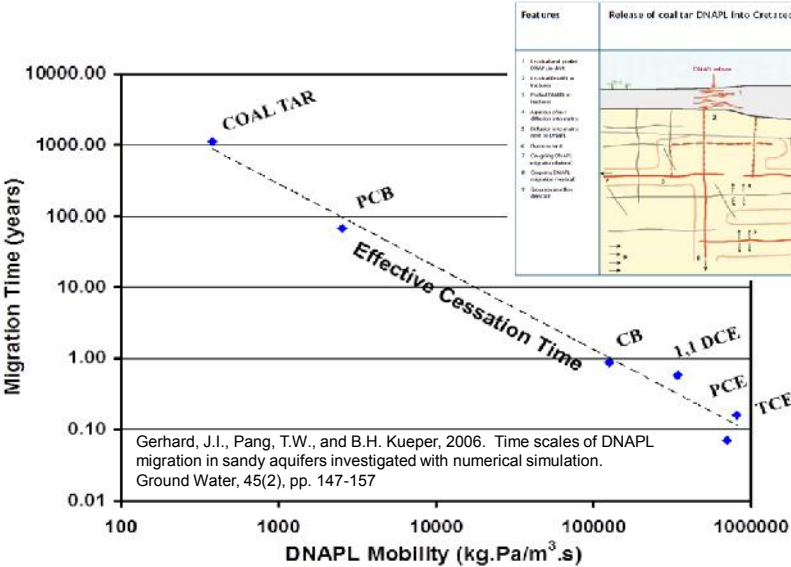
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DNAPL  
post spill stabilisation times



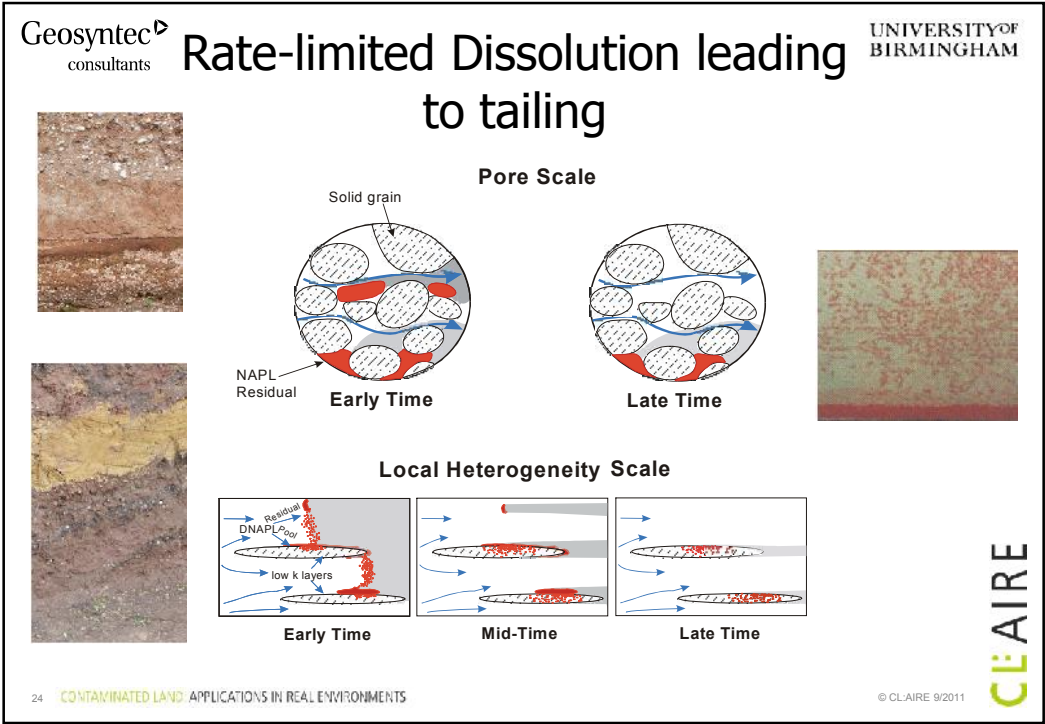
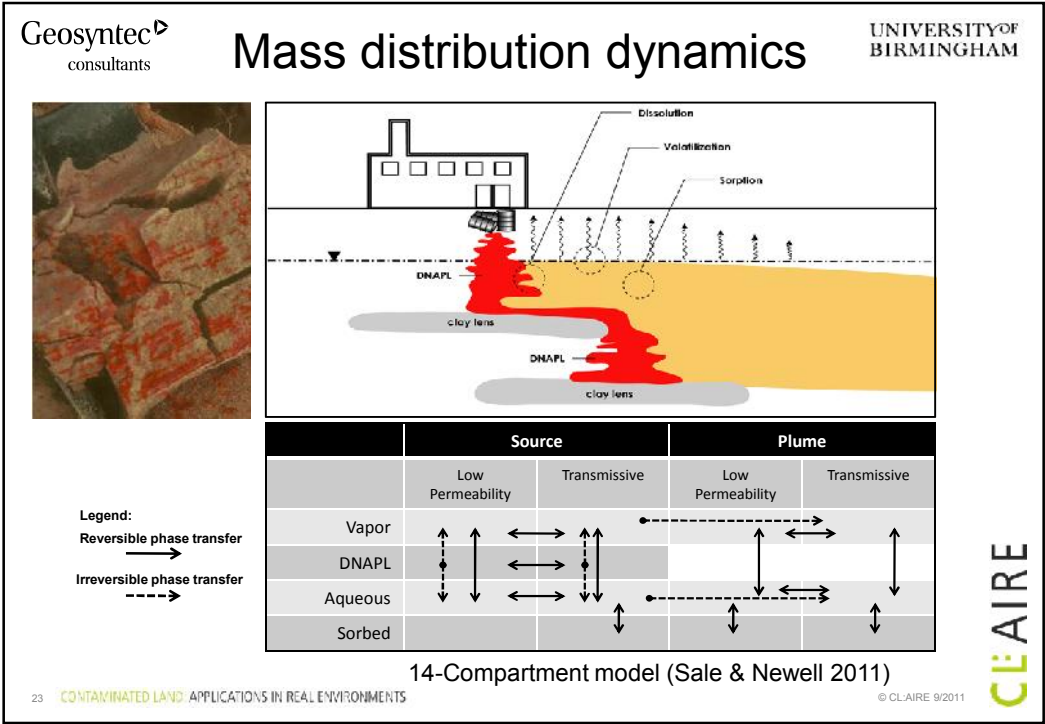
Features	Release of coal tar DNAPL into Cretaceous Chalk	Domain
1. Evaporation under 1000 Pa bar		Surface
2. Evaporation under 1000 Pa bar		Drift (unconsolidated)
3. Evaporation under 1000 Pa bar		Bedrock (unconsolidated)
4. Evaporation under 1000 Pa bar		Bedrock (unconsolidated)
5. Evaporation under 1000 Pa bar		Bedrock (unconsolidated)
6. Evaporation under 1000 Pa bar		Bedrock (unconsolidated)
7. Evaporation under 1000 Pa bar		Bedrock (unconsolidated)
8. Evaporation under 1000 Pa bar		Bedrock (unconsolidated)
9. Evaporation under 1000 Pa bar		Bedrock (unconsolidated)

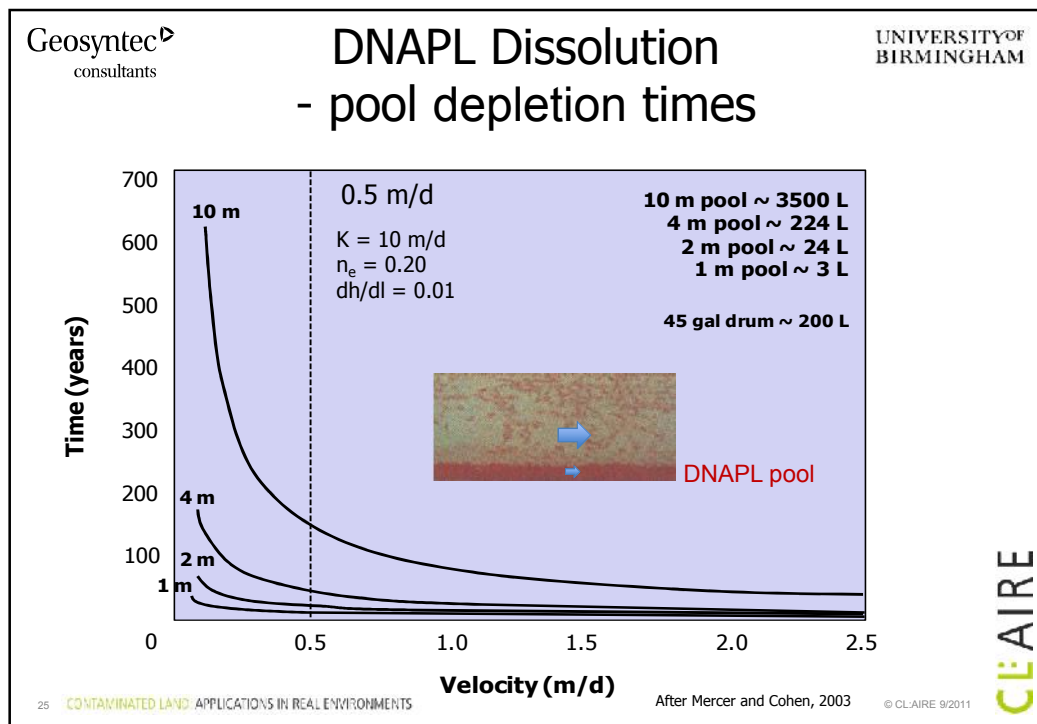
Gerhard, J.I., Pang, T.W., and B.H. Kueper, 2006. Time scales of DNAPL migration in sandy aquifers investigated with numerical simulation. Ground Water, 45(2), pp. 147-157

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## Solubility / Drinking water

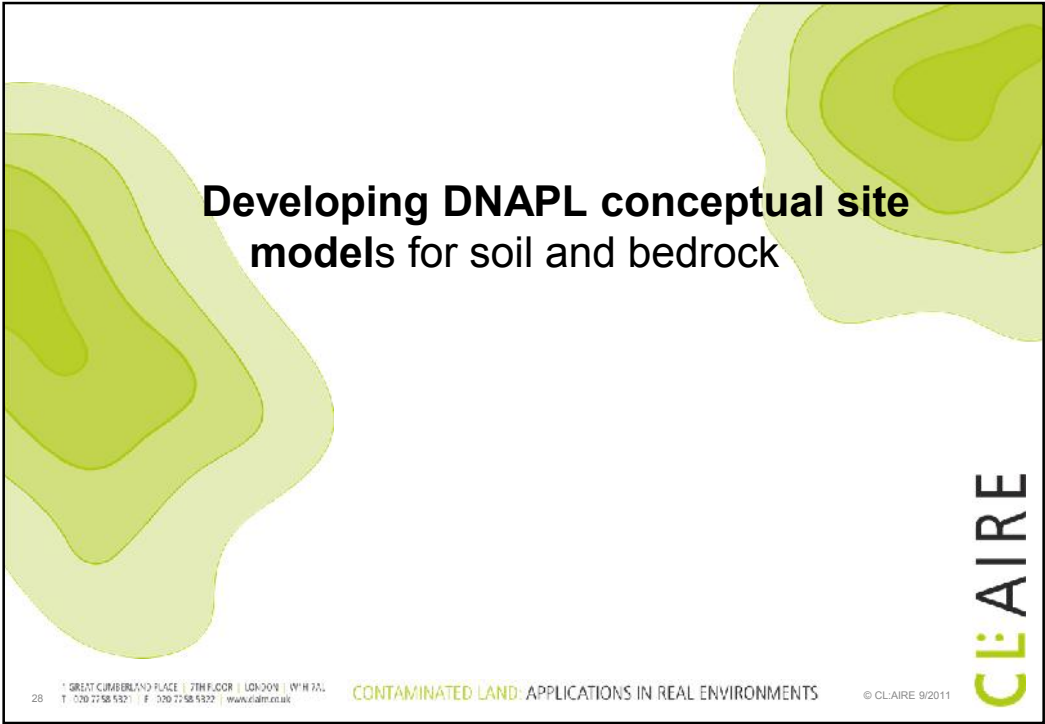
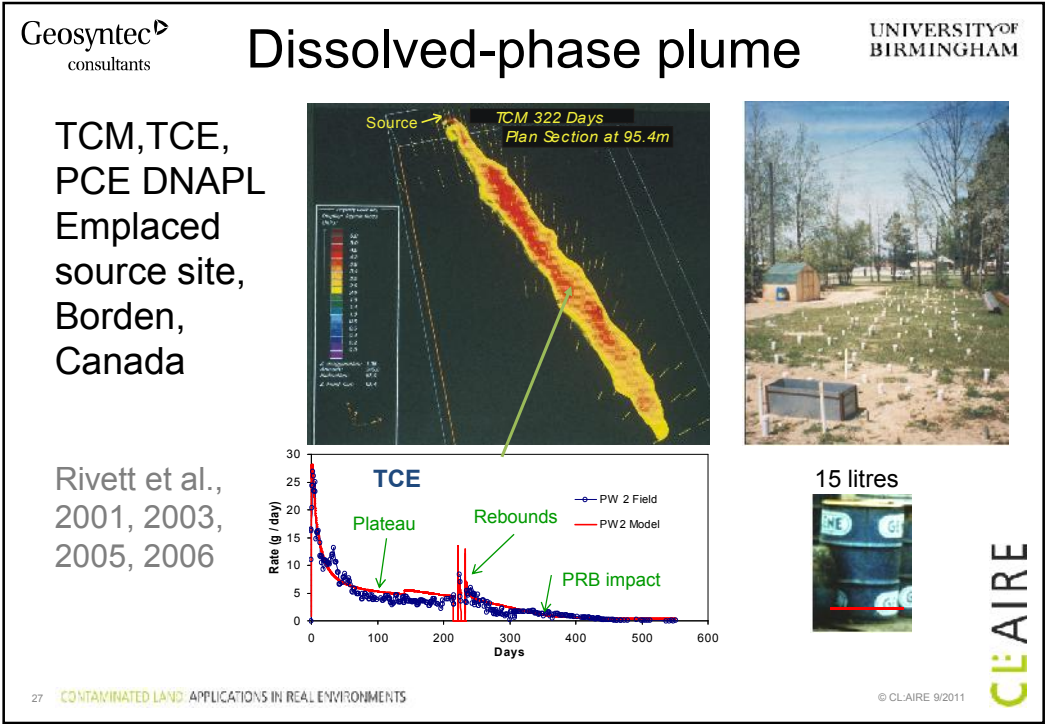
Chlorinated Organic Solvent	Solubility (mg/L)	MAC (mg/L)	Sol/MAC
1,2 Dichloroethane	8,690	0.003	$3 \times 10^6$
1,1,1-Trichloroethane	720	0.1	$7 \times 10^3$
CT	785	0.003	$2 \times 10^5$
Chloroform	8,200	0.2	$4 \times 10^4$
PCE	200	0.005	$4 \times 10^4$
TCE	1,100	0.005	$2 \times 10^5$

MAC is WHO maximum admissible concentration

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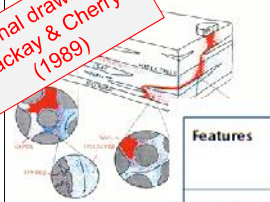
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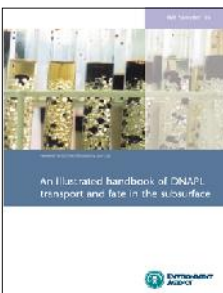
Original drawing of  
Mackay & Cherry  
(1989)

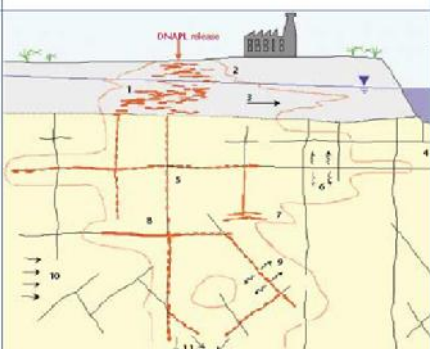


# Conceptual models

## The DNAPL paradigm shift

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Features	Release of chlorinated solvent DNAPL into Triassic Sandstone	Domain
1. Rejected and pooled DNAPL in drift		Surface
2. Vapor		Drift (unsaturated)
3. Aqueous phase plume migration		Drift (saturated)
4. Aqueous phase plume migration in fracture		Bedrock (saturated)
5. Residual DNAPL in fracture		
6. Aqueous phase matrix diffusion		
7. DNAPL penetration into coarse grained matrix		
8. Pooled DNAPL in fracture		
9. Matrix diffusion adjacent to DNAPL		
10. Groundwater flow direction		
11. DNAPL migration to depth		

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# Characterising DNAPL source zones, including direct and indirect tools, emerging investigation technologies and selection of which parameters to measure at DNAPL sites

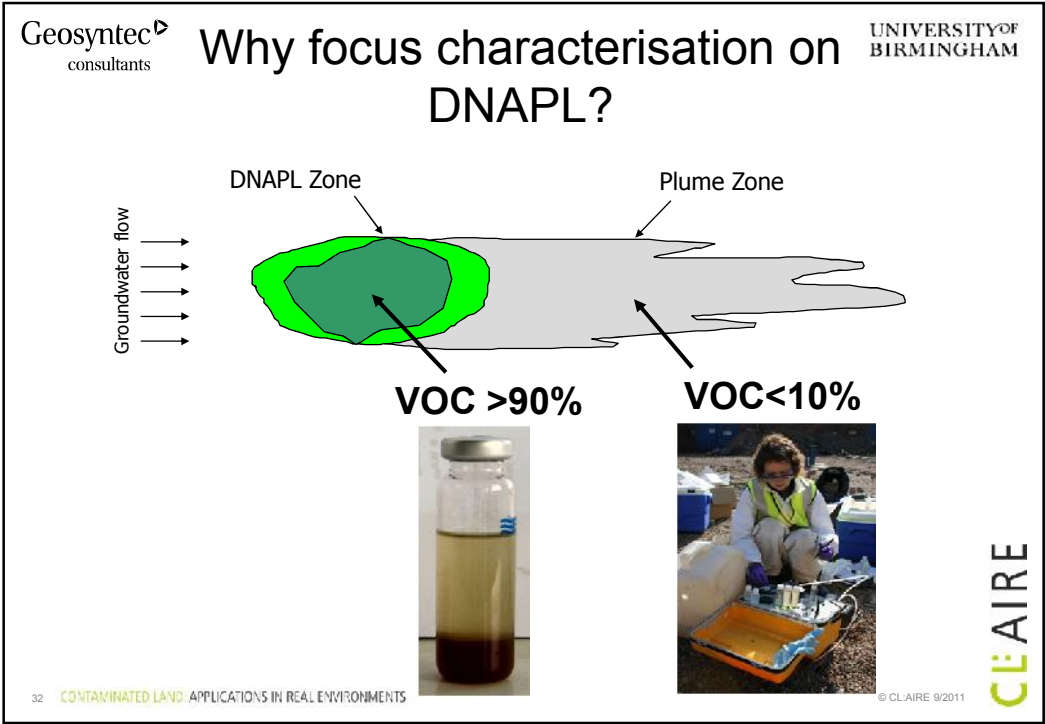
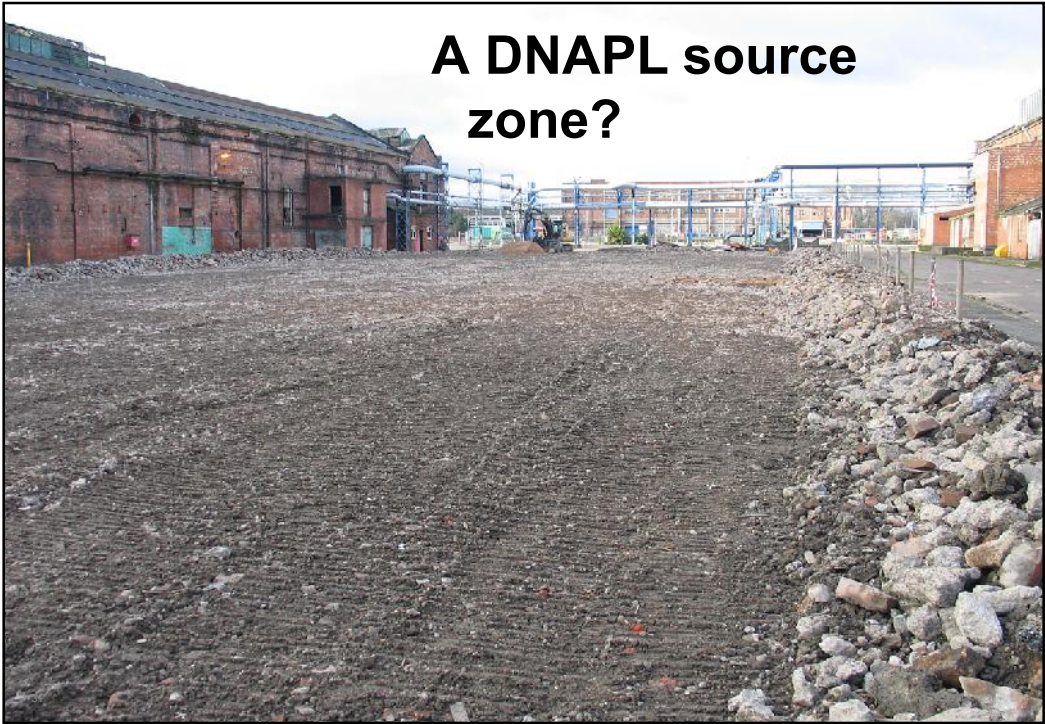
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## Multiple lines of evidence

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Direct measurement	Indirect measurement
<b>Visual observation</b>	<b>Site history</b>
<b>Enhanced visual methods</b>	<b>Soil gas survey</b>
<b>Drive point tools</b>	<b>Soil (partitioning threshold)</b>
	<b>Groundwater (Effective solubility)</b>
	<b>Partitioning tracer test</b>

- No SINGLE technique available to characterise a DNAPL site**
- Generally poor resolution, and resolution decreases with depth and hydrogeological complexity**
- Resolution typically constrains selection, application and performance assessment of remediation technologies**

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## Direct: Visual observation

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**From core or grab samples**


- Rare at chlorinated solvent sites
- Common at coal tar sites

\* IFT = 7 dynes/cm  
\* Density ≈ 1.4 g/cm<sup>3</sup>  
\* Contact angle 30°

**Samples recovered during from pumping from wells**

- Typically requires pooled DNAPL

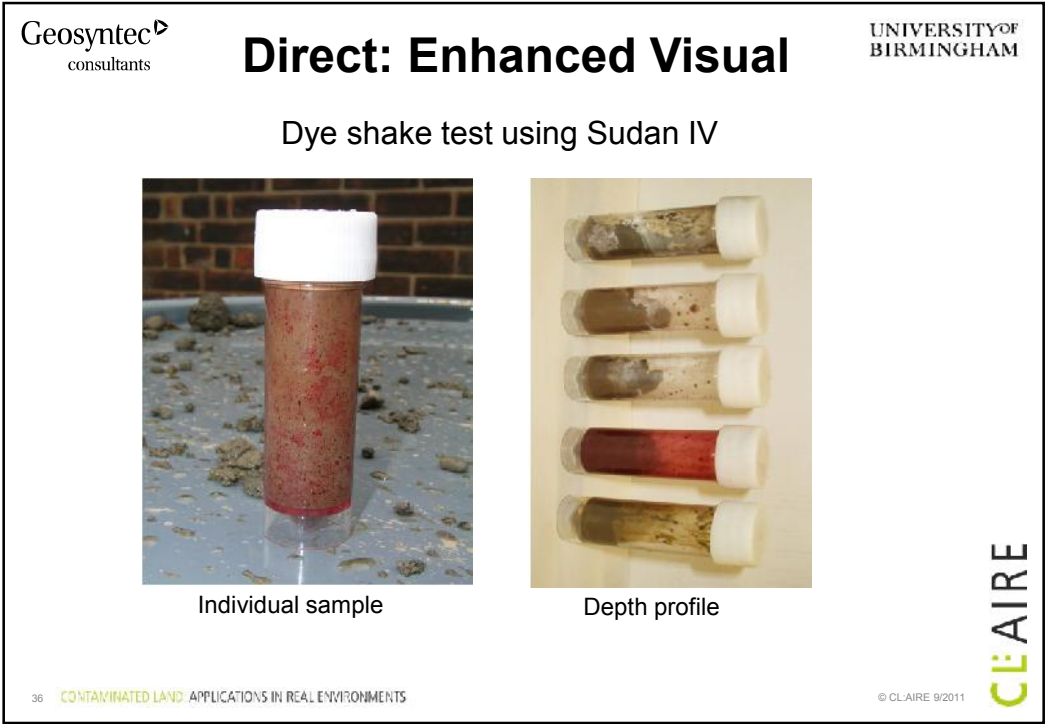
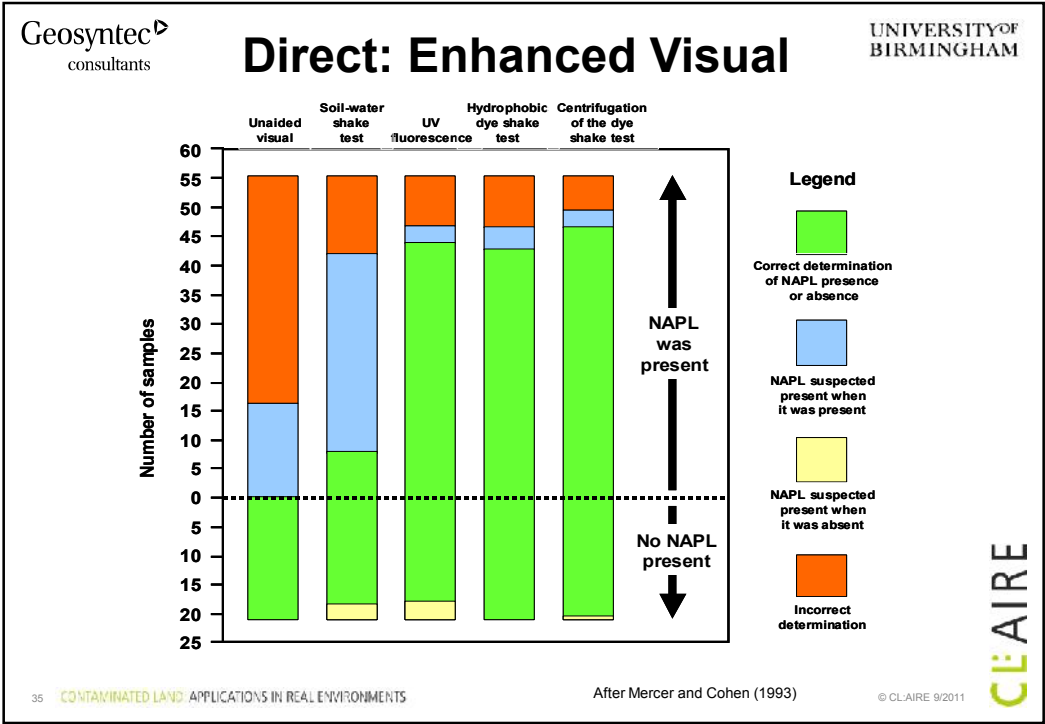


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Direct: Enhanced visual

Flexible liner underground everting membrane [Flute]

tubing to bottom of liner  
carrier liner  
reactive covering  
water fill in liner  
surface  
stain on cover  
DNAPL pond on stratum  
rod tip left in hole

First rods driven into ground  
liner lowered into rods  
rods raised with water inside liner  
"the trick"... liner dilates in hole, but not in rod  
result, water filled liner in hole  
liner and cover removed from hole  
tubing used to invert liner from hole  
reactive cover pressed against hole wall by water filled liner  
Sequence of steps in procedure

From: [http://www.flut.com/sys\\_3.html](http://www.flut.com/sys_3.html)

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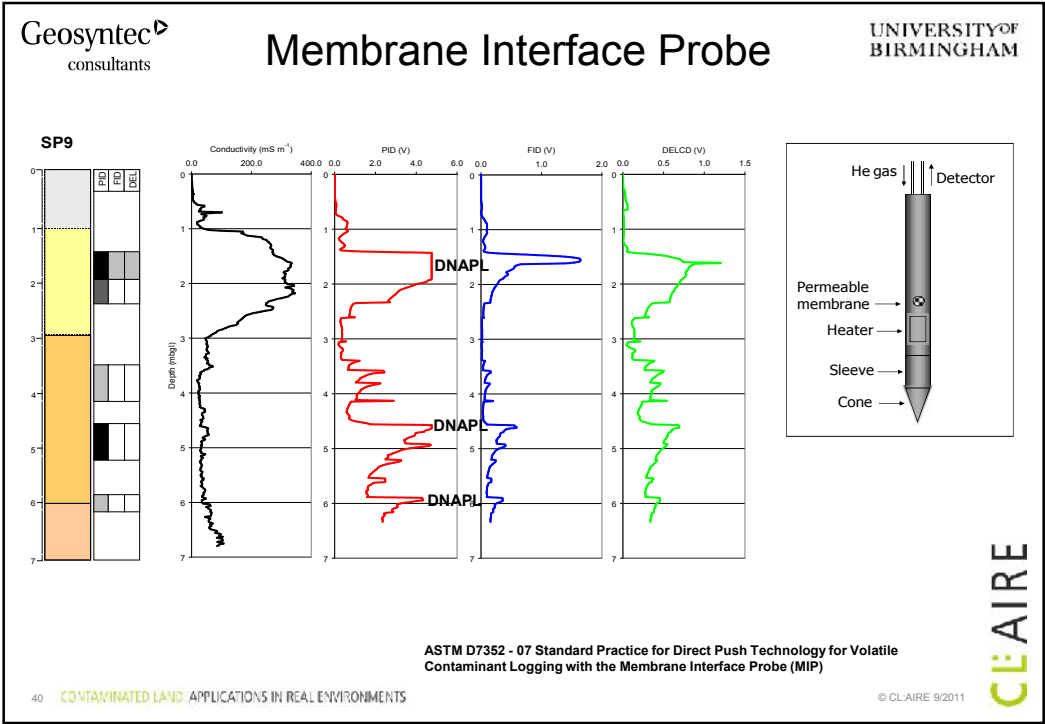
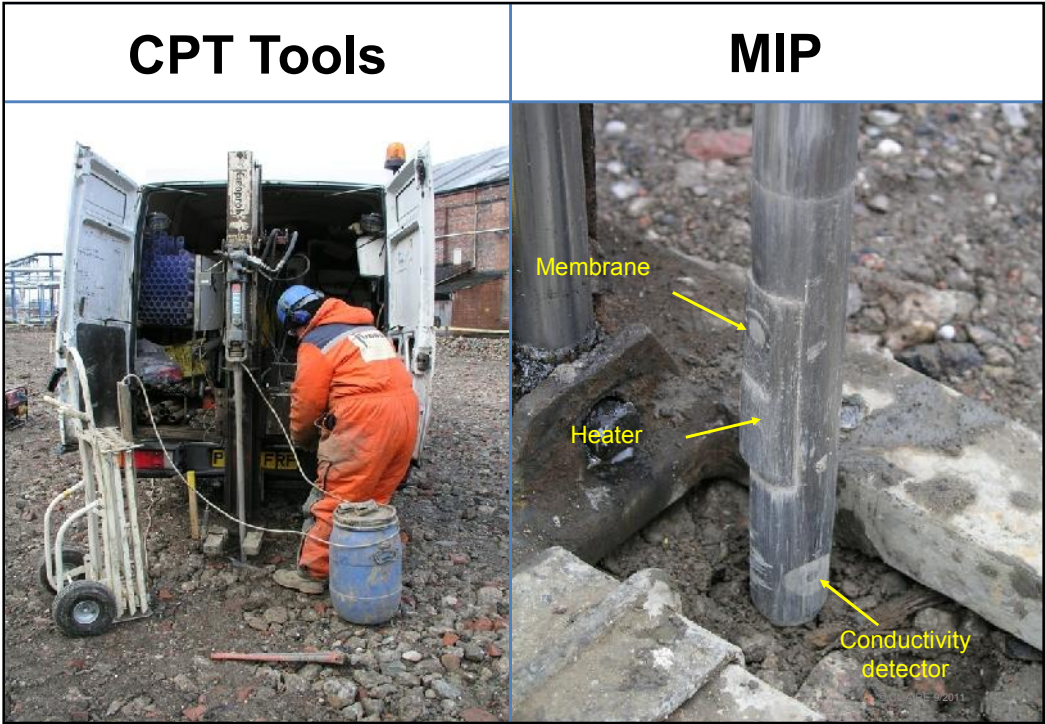
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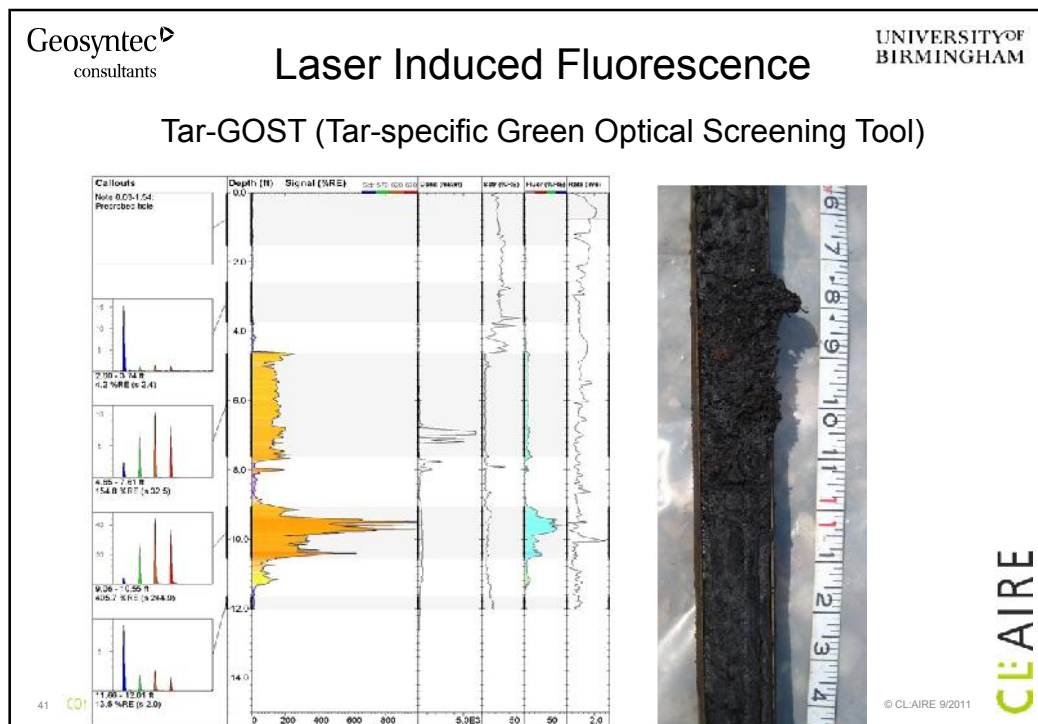
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Direct: Drive Point Tools

Method	Advantages	Disadvantages
Video imaging [GeoVis]	<ul style="list-style-type: none"><li>Can be coupled with lithological logs</li><li>Data easy to interpret in suitable soil matrix</li></ul>	<ul style="list-style-type: none"><li>Limited by lithology</li><li>Transparent DNAPL not visible</li><li>Clays may smear the camera window</li></ul>
Membrane Interface Probe [MIP]	<ul style="list-style-type: none"><li>Can be coupled with lithological logs</li><li>Range of detectors [FID, PID, ECD]</li><li>Excellent screening level data</li></ul>	<ul style="list-style-type: none"><li>Limited by lithology</li><li>Semi-quantitative</li><li>High operator skill required</li></ul>
Laser Induced fluorescence [LIF]	<ul style="list-style-type: none"><li>Can be coupled with lithological logs</li><li>Simple detection methodology</li><li>Excellent screening level data</li></ul>	<ul style="list-style-type: none"><li>Limited by lithology</li><li>Semi-quantitative</li><li>False positives may be detected</li></ul>
Dye-LIF	<ul style="list-style-type: none"><li>Can be coupled with lithological logs</li><li>Applicable to non-fluorescing compounds</li><li>Excellent screening level data</li></ul>	<ul style="list-style-type: none"><li>Limited by lithology</li><li>Still under development</li></ul>
TarGOST	<ul style="list-style-type: none"><li>Can be coupled with lithological logs</li><li>Applicable to coal tar compound</li><li>Excellent screening level data</li></ul>	<ul style="list-style-type: none"><li>Limited by lithology</li><li>Only applicable to coal tar compounds</li></ul>

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## Indirect: Site history

- Previous investigations
- Aerial photographs
- Building plans
- Former lagoons, USTs, drains
- Production records
- Employee interviews
- Sale records


1954

2011

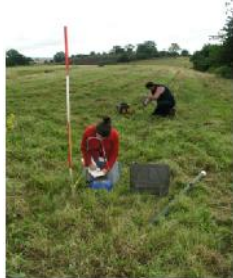
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## Indirect: Soil gas survey




- Vapour phase plume may be coincident with DNAPL source zone
- Avoid mistaking vapours from a groundwater plume from vapours derived from DNAPL
- Requires other supporting lines of evidence
- Methods include shallow soil spike survey or sampling monitoring wells (active or passive)
- Radon-222 has been used to infer DNAPL presence




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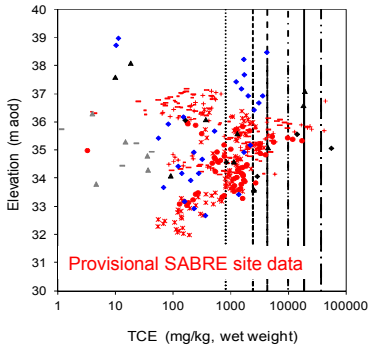
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## Indirect: Soil partitioning threshold



$$C_D = \frac{S_r \phi \rho_N 10^6}{\rho_b} + C^T$$




Provisional SABRE site data

x ST76 (T1) 6 m  
 • ST77 (T1+T4) 6 m  
 - ST78 (T1) 6 m  
 + ST79 (T1) 6 m  
 - ST80 (T1) 6m  
 • SF45 (T2) 10 m  
 • SF46 (T2) 10 m  
 • IW107 (T4) 13 m  
 • SF52 (T3) 23 m  
 - SF53 (T3) 23 m  
 ..... DNAPL threshold  
 - - - DNAPL sat = 1%  
 - - - DNAPL sat = 2%  
 - - - DNAPL sat = 5%  
 - - - DNAPL sat = 10%  
 - - - DNAPL sat = 20%

Where:  
 $C_D$  is soil concentration (mg/kg) corresponding to threshold DNAPL saturation  
 $S_r$  is threshold DNAPL saturation  
 $\phi$  is the porosity  
 $\rho_N$  is DNAPL density (g/cc)  
 $\rho_b$  is dry soil bulk density (g/cc)  
 $C^T$  is amount of contaminant (mg/kg) in the samples, including aqueous, vapour and sorbed  
  
 Kueper and Davies, 2009

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## Indirect: Theoretical pore water concentration

### 1) Determine theoretical pore water concentration



$$C_w = \frac{C_t \rho_b}{(K_d \rho_b + n)}$$

$C_w$  is the pore water concentration (mg/L)  
 $C_t$  is the soil concentration (mg/kg)  
 $\rho_b$  is the bulk density (g/cm<sup>3</sup>)  
 $K_d$  is the partition coefficient (cm<sup>3</sup>/g)  
 $n$  is porosity (dimensionless)

### 2) Compare theoretical pore water concentration to the solubility ( $S$ ) or effective ( $S_e$ ) of the compound of interest

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Feenstra et al., 1991

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## Inferred DNAPL presence

US EPA guidance indicates that  
**1% effective solubility** infers  
the presence of DNAPL

$$S_i^e = X_{mi} S_i$$

Where:

$S_i^e$  is the effective solubility of component  $i$  in water  
 $X_{mi}$  is the mole fraction of component  $i$  in a given mixture  
 $S_i$  is the solubility of component  $i$  in water



**Consider concentration trends with depth**

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Inferred DNAPL presence

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Chlorinated Organic Solvent	Solubility (mg/L)	1% (mg/L)
1,2 Dichloroethane	8,690	87
1,1,1-Trichloroethane	720	7
CT	785	8
Chloroform	8,200	82
PCE	200	2
TCE	1,100	11

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Groundwater sampling  
downgradient of a DNAPL source zone

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DNAPL release

Groundwater flow

DNAPL

dissolved plume

5 mg/l, 35 mg/l, 3 mg/l, N.D., 1 mg/l, N.D.

75% mass flux discharges through 5-10% plume cross-sectional area (Guilbeault, et. al., 2005)

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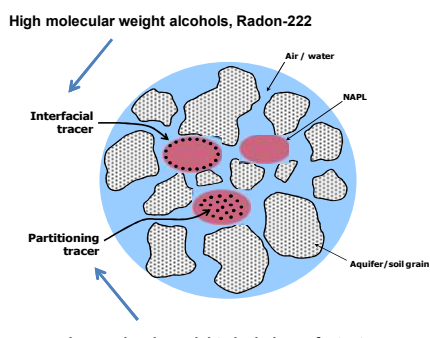
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## Indirect: Partitioning and interfacial tracers

**NAPL morphology index ( $H_{NW}$ )** proposed to determine the expected efficiency of remediation by in situ flushing techniques :

$$H_{NW} = \alpha_{NW} / \phi S_{NW}$$

Where:  $\phi S_{NW}$  is the partitioning tracer and  $\alpha_{NW}$  is the interfacial tracer



- **High values of  $H_{NW}$**  indicate a source that is characterised by **RESIDUAL DNAPL**
- **Low values of  $H_{NW}$**  indicate a source that is characterised by **POOLED DNAPL**

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## What parameters should be measured?

### Contaminant characteristics

Parameter	Example use of information
DNAPL density	DNAPL mobility and pool height calculations
DNAPL viscosity	Determine if DNAPL could still be moving Design of NAPL recovery system
DNAPL component composition	Effective solubility calculations Predict future composition of plume
DNAPL-water interfacial tension	Determine importance of capillary forces Pool height calculations
Organic carbon partition coefficient	Determine degree of aqueous phase sorption and rate of plume migration
Contaminant half-life	Determine degree of degradation and rate of plume migration
DNAPL vapour pressure	Determine if vapour migration is a potential issue; Estimate lifespan of DNAPL above water table
Date and volume of DNAPL release	Estimate of depth of DNAPL migration. Is DNAPL still moving?
Potential DNAPL release locations	Help guide monitoring well placement

From Illustrated handbook of DNAPL transport and fate in the subsurface (Environment Agency, 2004)

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## What parameters should be measured?

### Drift (overburden) characterisation

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Parameter	Example use of information
Porosity	Plume velocity calculation; Diffusion calculations
Dry bulk density	DNAPL threshold concentration calculation
Fraction organic carbon	Plume velocity calculation; DNAPL threshold calculation
Hydraulic conductivity	Plume velocity calculation; Design of extraction wells
Displacement pressure	Pool height calculations
Bulk retention capacity	DNAPL mass estimate
Contact angle	Refinement of conceptual model on DNAPL mobility
Hydraulic head distribution	Directions of groundwater flow and velocity of groundwater
Bedding structures	Directions of DNAPL migration
Spatial extent of DNAPL source zone	Guide remedy selection and design
Spatial extent of plume	Guide remedy selection; risk analysis

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From **Illustrated handbook of DNAPL transport and fate in the subsurface**  
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## What parameters should be measured?

### Bedrock characterisation

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Parameter	Example use of information
Matrix porosity	Diffusion calculations
Matrix dry bulk density	Estimate of remediation timeframe
Matrix fraction organic carbon	Estimate of (retarded) plume velocity
Orientation of major fracture sets	Determine direction of plume migration Directions of DNAPL migration
Fracture spacing	Diffusion calculations
Fracture porosity	Plume velocity calculation
Bulk rock hydraulic conductivity	Plume velocity calculation Design of extraction wells
Hydraulic head distribution	Directions of groundwater flow and velocity of groundwater
Bulk retention capacity	DNAPL mass estimate
Contact angle	DNAPL-rock-water wetting relationship
Spatial extent of DNAPL source zone	Guide remedy selection
Spatial extent of plume	Guide remedy selection; risk analysis

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**Calculating DNAPL mass estimates**  
from typical site investigation data.  
Understanding and communicating  
uncertainty in contaminant mass  
distribution to stakeholders.

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**Site characterisation for  
DNAPL mass estimation**

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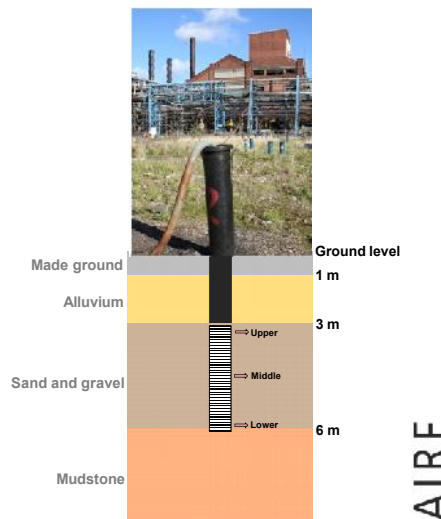
1. Groundwater monitoring from fully  
screened monitoring wells to  
determine pre- and post-treatment  
DNAPL mass
2. Detailed (research level) site  
characterisation to determine pre- and  
post-treatment DNAPL mass

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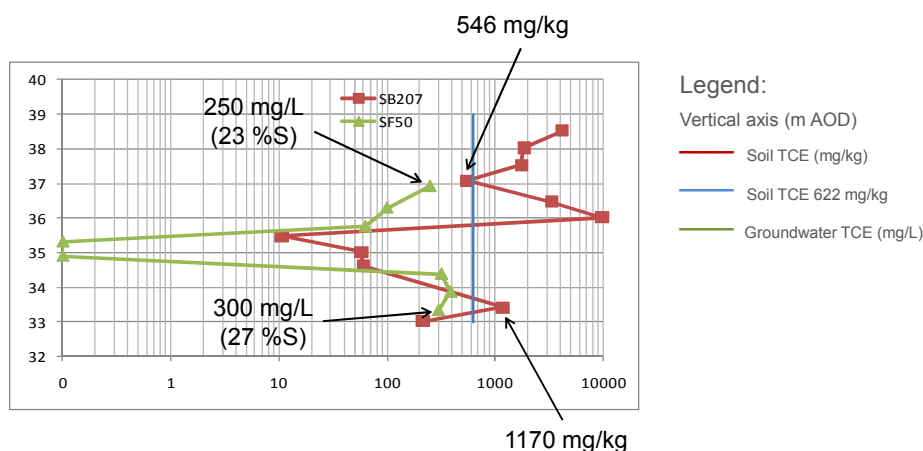
## 1. Groundwater monitoring data

- 1) Groundwater samples analysed for TCE from three levels in fully screened wells
- 2) Determine volume of aquifer where groundwater concentration is above a given solubility threshold (equivalent to DNAPL)
- 3) Estimate DNAPL mass using the bulk retention capacity expression

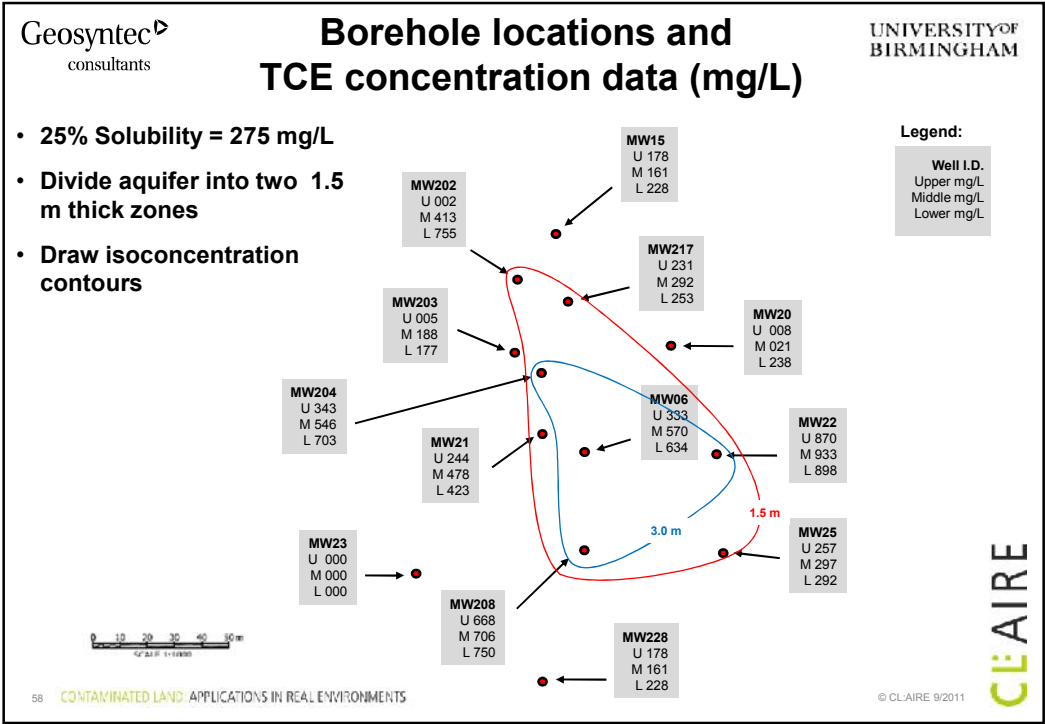
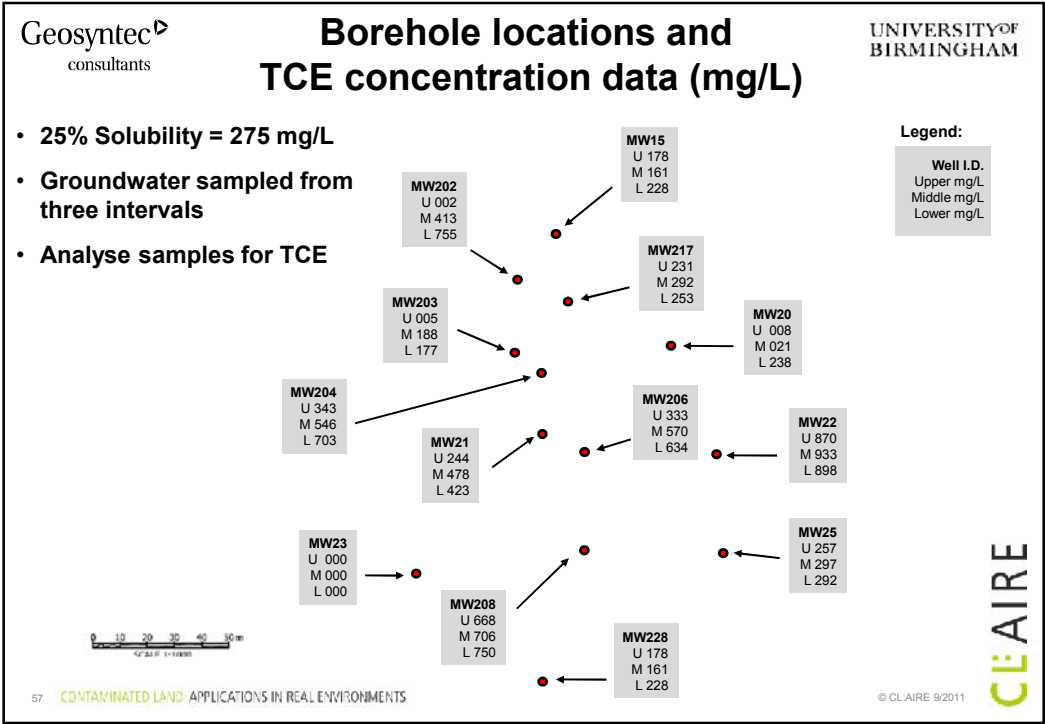


## Solubility threshold

**Site soil and groundwater data indicated that 275 mg/L (25% of solubility) was representative of DNAPL**



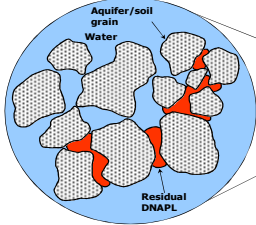




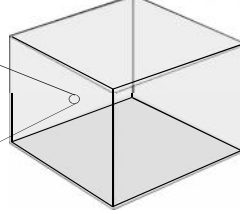
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## DNAPL mass estimation

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**Residual saturation**  
Proportion of pore space occupied by DNAPL



**Bulk retention capacity**  
Proportion of a given volume of the subsurface occupied by DNAPL

$$M_{DNAPL} = \sum S_r \theta V \rho_{TCE}$$

Where:  $S_r$  is the residual non-wetting phase (DNAPL) saturation,  $\theta$  is fractional porosity,  $V$  is the volume of aquifer, and  $\rho$  is the density of the DNAPL

$\theta = 0.30$   
 $S_r = 0.74\%$   
 $\rho = 1460 \text{ kg/m}^3$   
 **$V$  = volume determined from groundwater sampling**

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## Inferred DNAPL mass estimates from groundwater sampling

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Depth range	Total area	Total volume	Total porosity	Total $S_r$ volume	Total TCE mass
m	m <sup>2</sup>	m <sup>3</sup>	m <sup>3</sup>	m <sup>3</sup>	kg
3.0-6.0	3,675	11,025	3,308	24.48	35,734
4.5-6.0	4,375	6,563	1,968	14.57	21,270
<b>Total</b>	<b>8,050</b>	<b>17,588</b>	<b>5,276</b>	<b>39</b>	<b>57,005</b>

**TCE DNAPL mass ≈ 57 tonne**

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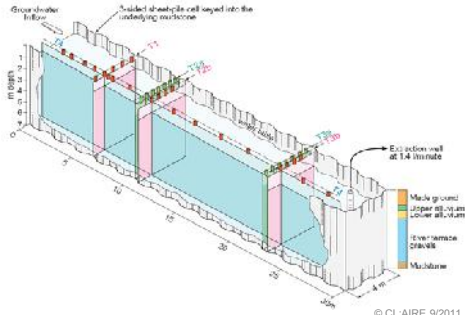
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# The SABRE project

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- Bioremediation research project
- Project involved highly detailed characterisation of a DNAPL source zone
- Aim to quantify mass depletion and assess uncertainty in DNAPL mass estimates



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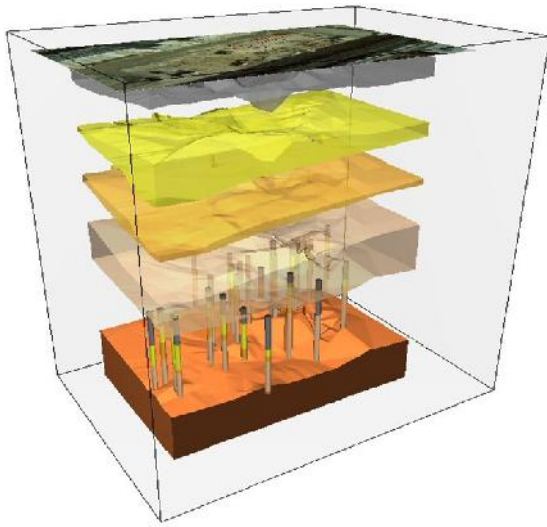
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# Geological reconstruction

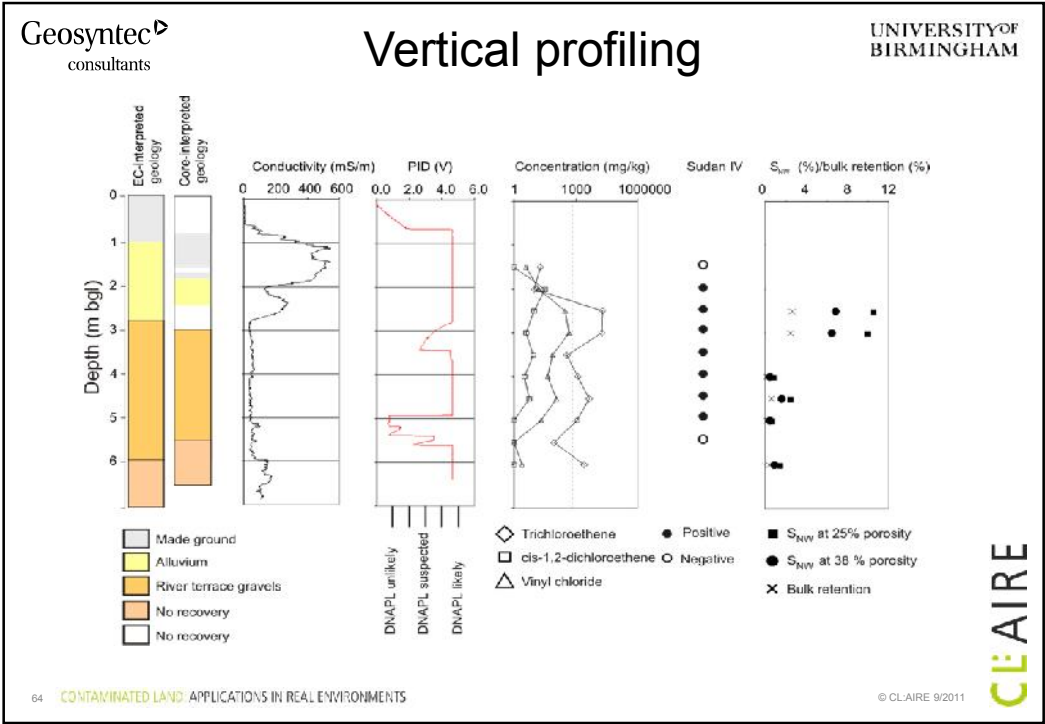
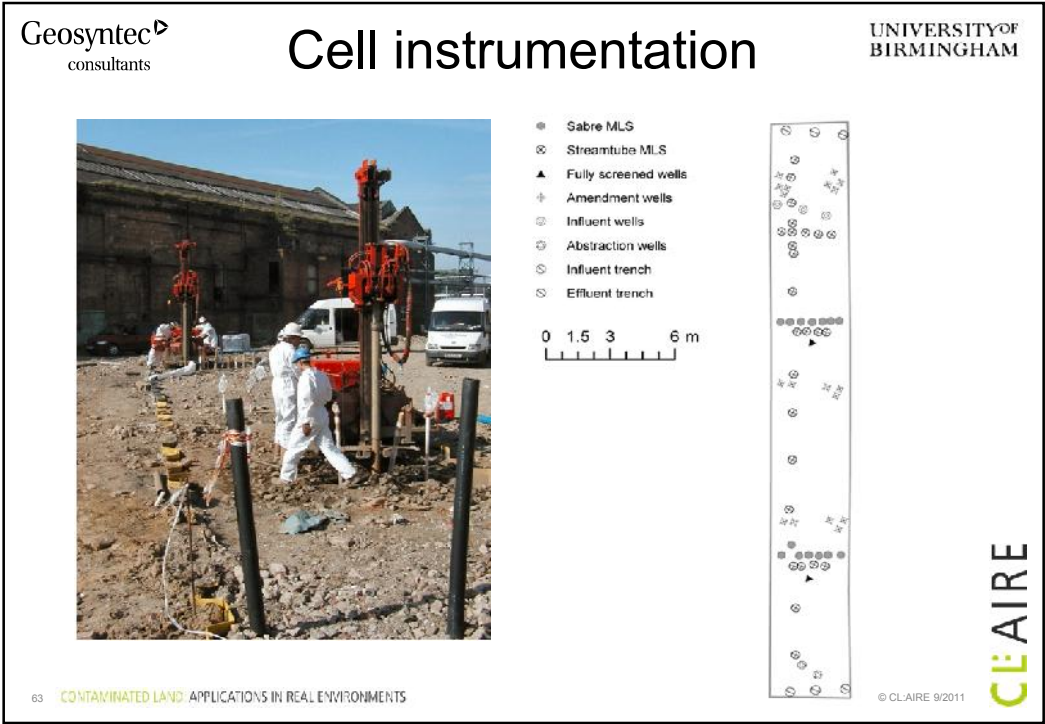
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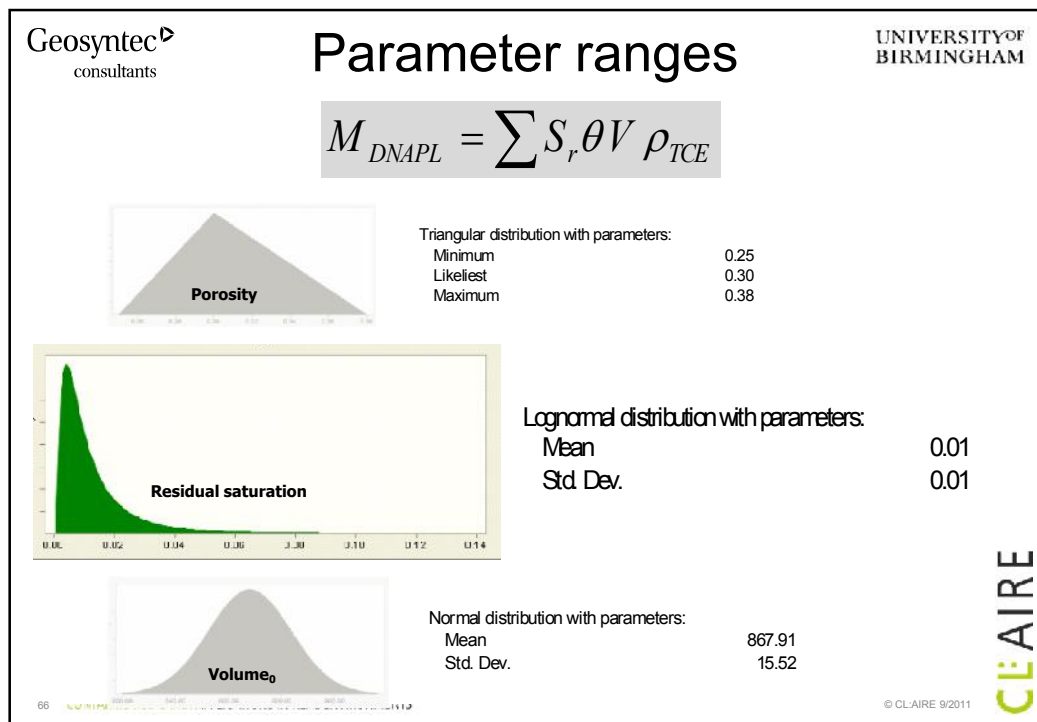
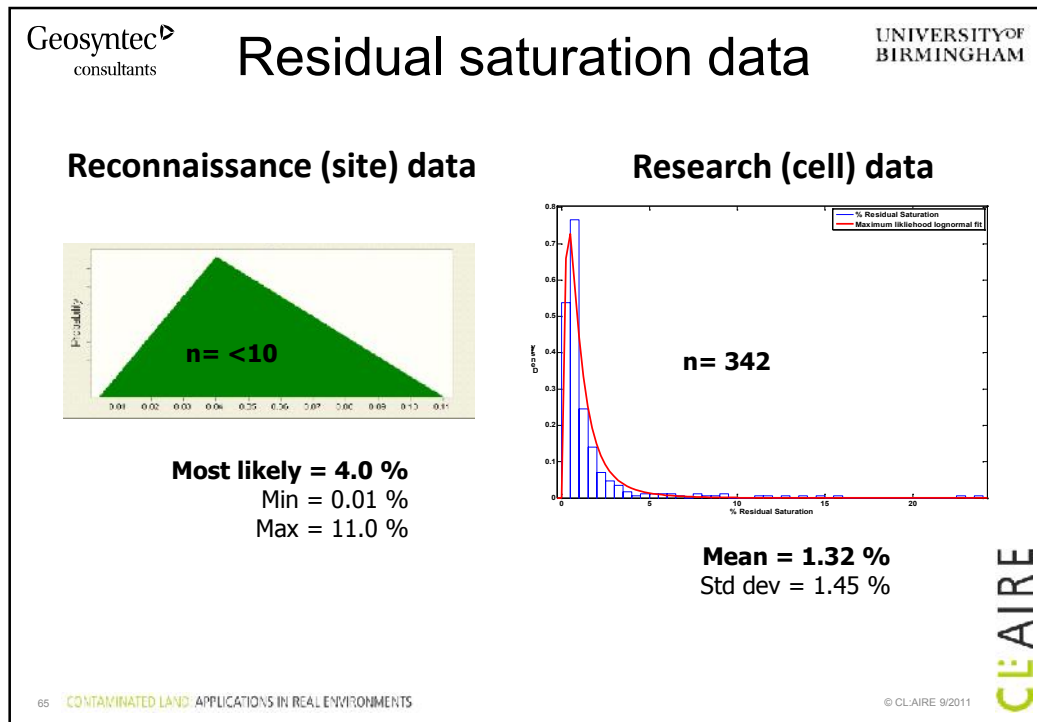


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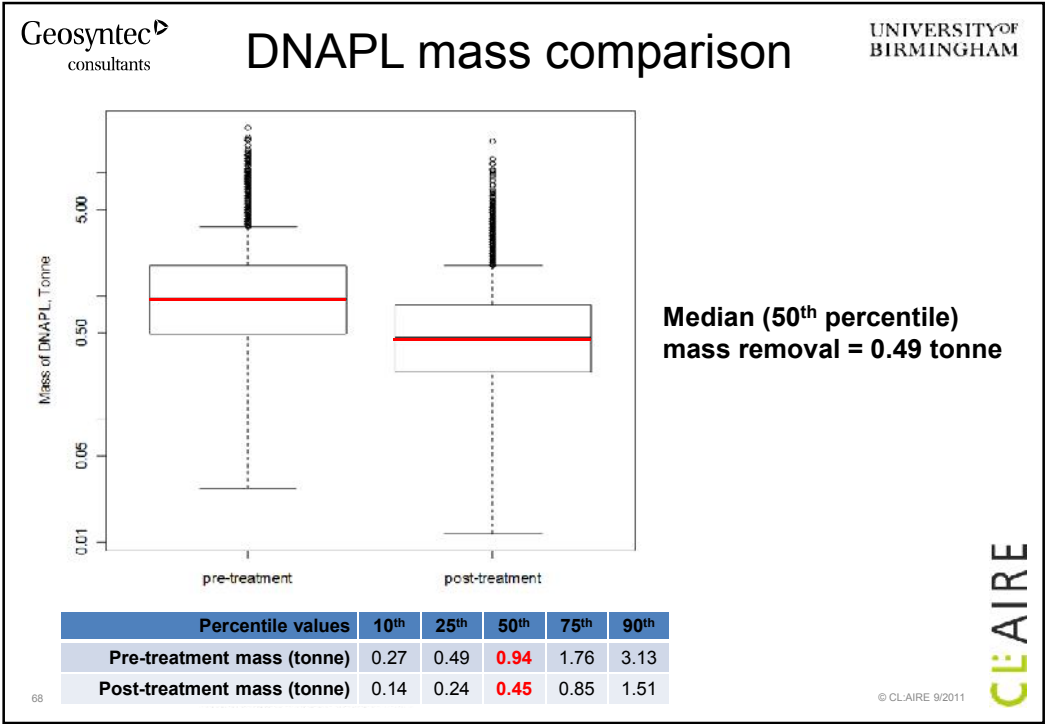
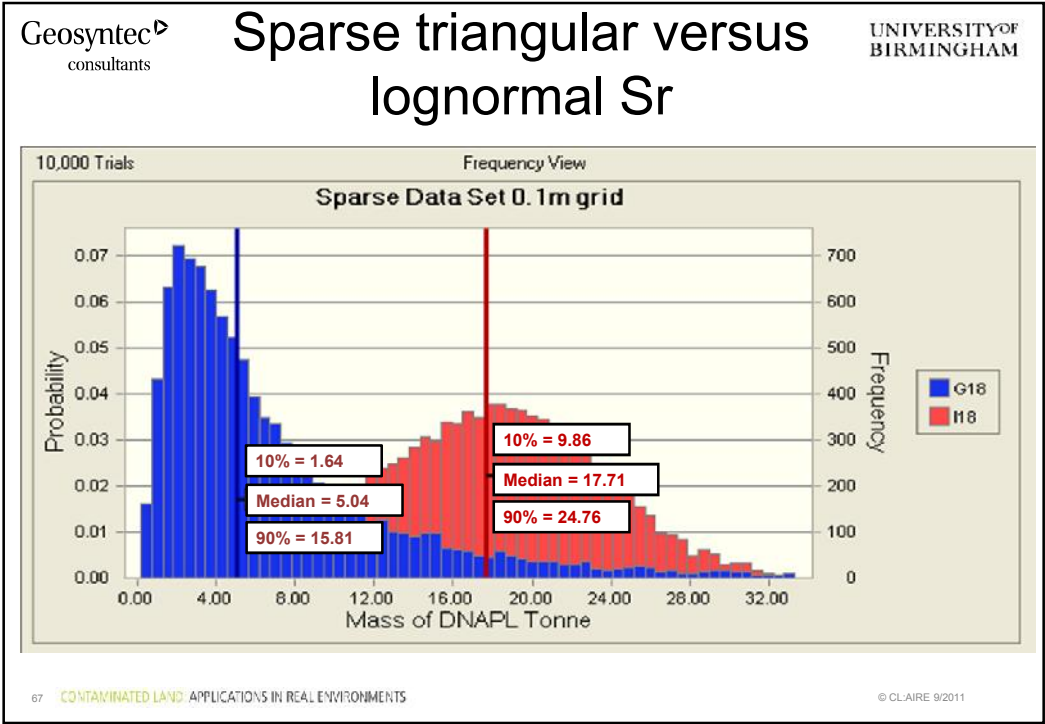
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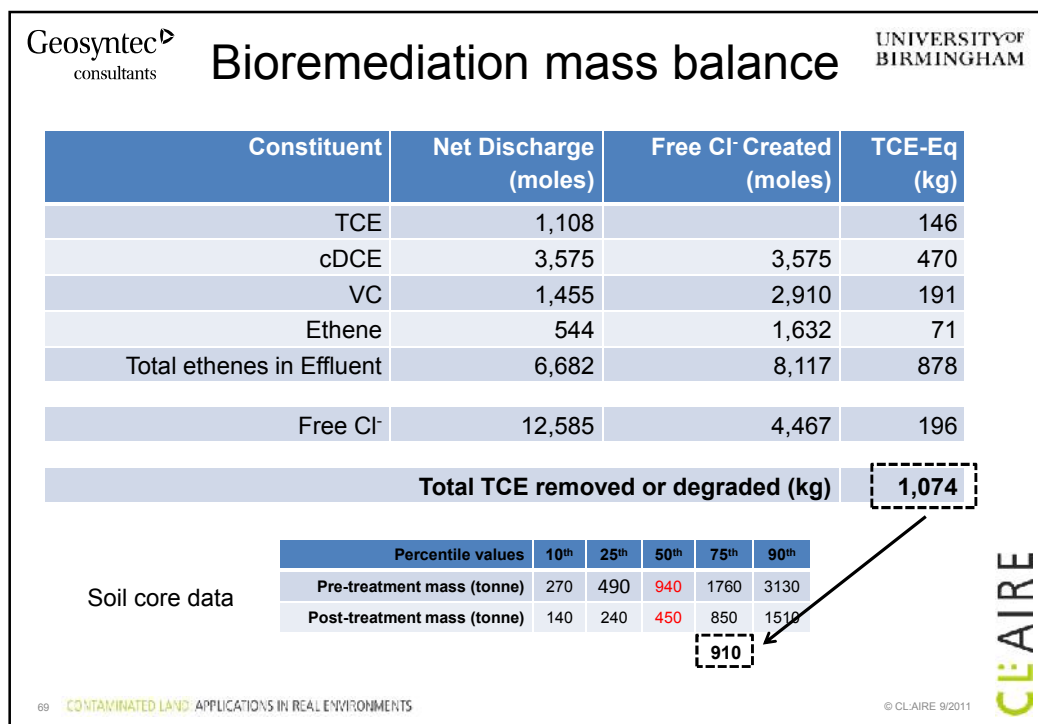
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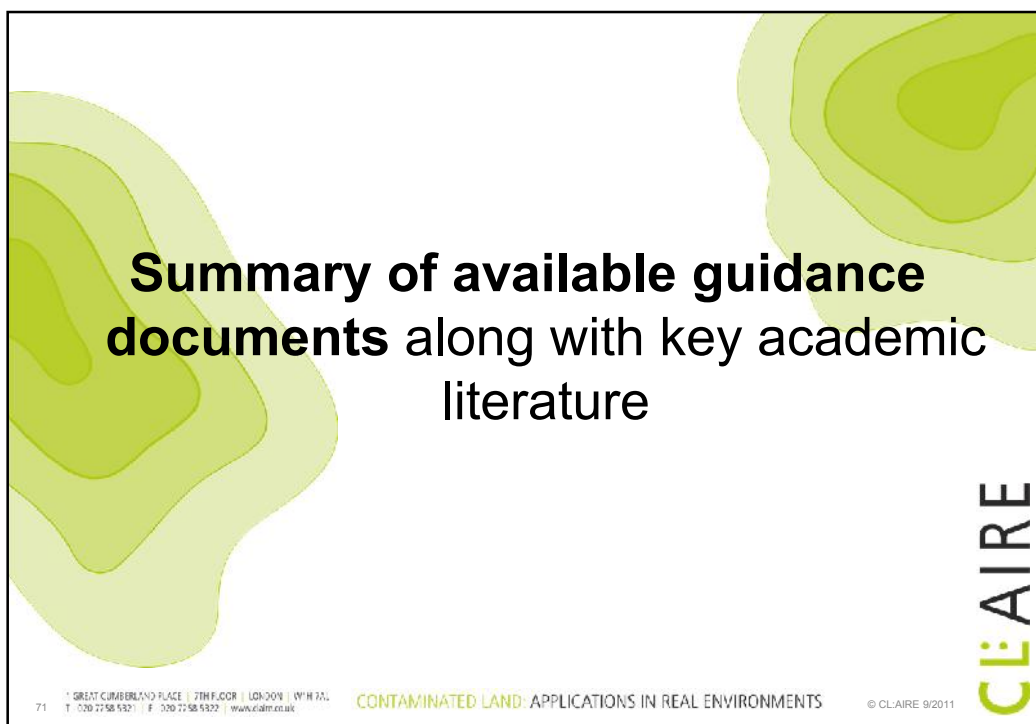
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## Challenges

- High spatial measurement density required for 3-D characterisation of DNAPL sources
- Improved resolution of high mass density zones – pools, low permeability zones
- Assessment methods that are rapid, cost-effective, suited to bedrock geologies
- Improved predictability of DNAPL source zone longevity and temporal flux reductions
- Estimation of DNAPL mass diffused to immobile zones and predicted temporal release

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## Selected literature



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## Useful literature listing

*Cited and Useful literature listing*




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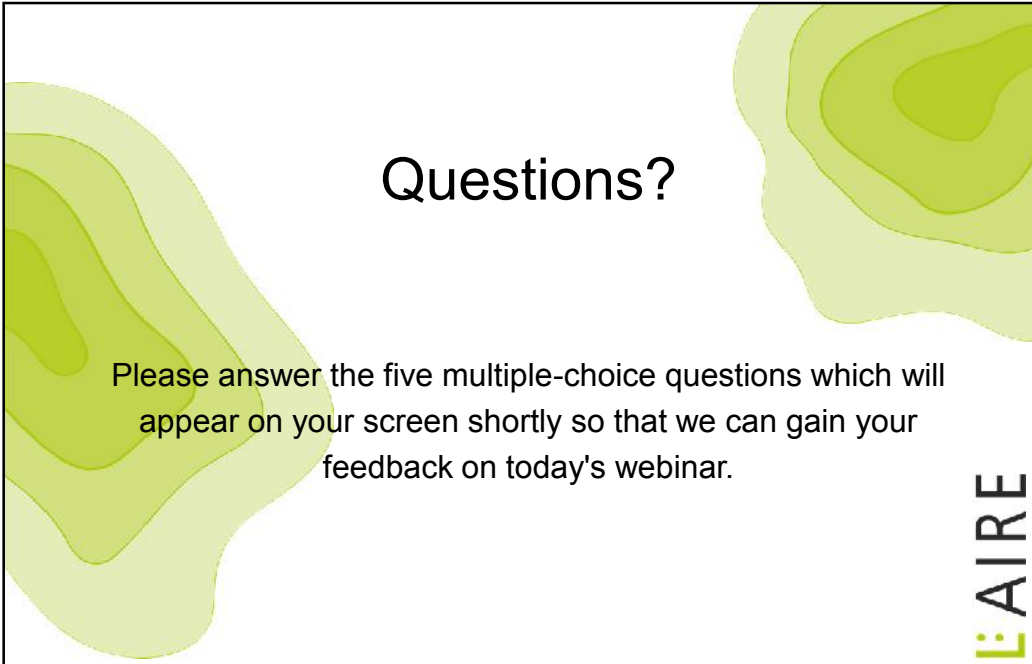
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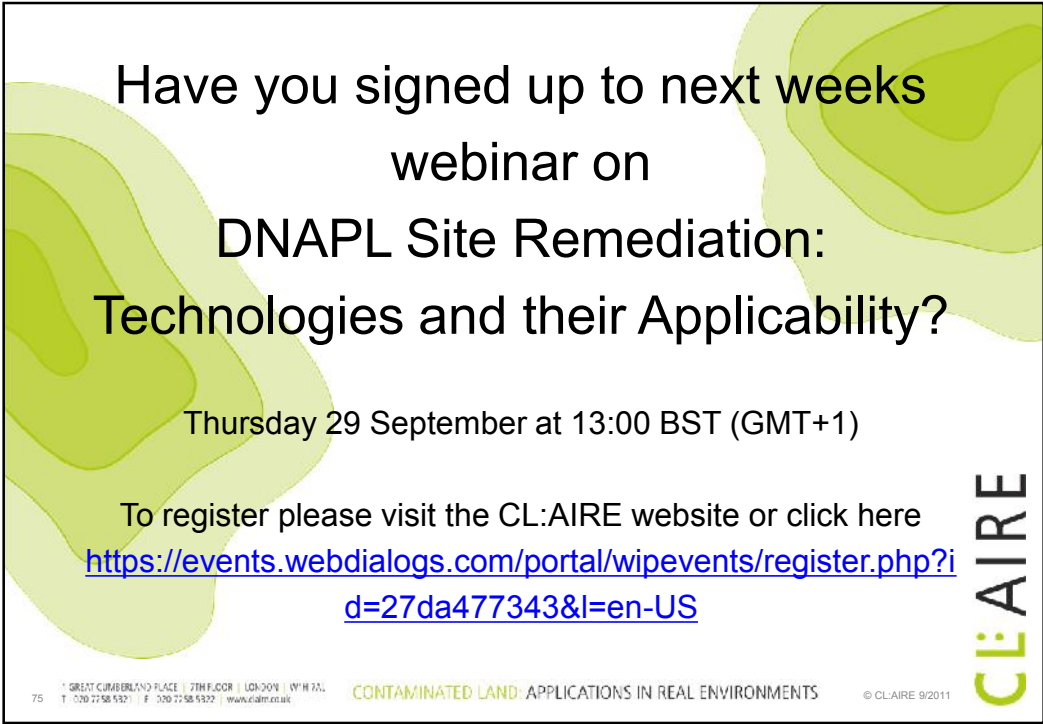


## Questions?

Please answer the five multiple-choice questions which will appear on your screen shortly so that we can gain your feedback on today's webinar.



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