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Assessing Risk from DNAPLs in Fractured Aquifers

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تقييم المخاطر الناجمة من السوائل الكثيفة غير مائية الطور (DNAPLS) في الأحواض المتصدعة

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خلاصة: تعد المذيبات المكلورة من ضمن أهم ملوثات المياه الجوفية. وتتحرك السوائل الكثيفة غير مائية الطور (DNAPLS) والتي تنساب بسرعة وبشكل معقد في الشقوق للوصول إلى الأحواض الجوفية ومن ثم تلويث أجزاء كبيرة منها ومن ثم تذوب لتتسبب في تلوث المياه الجوفية بشكل خطير. إن التخلص من هذه السوائل ومن الصخور المتصدعة يعتبر أمرا مستحيلا وان أمكن فهو باهظ الثمن ويجب تقييم المخاطر الناجمة عن التلوث بهذه السوائل لمعرفة مستوى التلوث ولقيام بتدابير تنظيف أو منع انتشار التلوث. تهدف هذه الدراسة الى معرفة مدى أو عمق انسياب هذه السوائل في شبكة التصدعات الصخرية وتناقش هذه الورقة قياس سعة التصدعات في الموقع وترابط شبكات الشقوق بالنسبة لهذه السوائل. لقد تم تطوير وتطبيق تقنية حقلية لقياس تباين سعة الشقوق وضغط دخول هذه السوائل في الصخور المتشققة والمشبعة بالماء. لقد أوجدت التجربة الحقلية أيضا الفرصة لقياس نفاذية الوجه الرطب عند الوجه المشبع الغير رطب. إن تقنية هذه السوائل تعمل على ضخ معدل ثابت منها غير سام في صدع أو شق معزول ومحمي بطبقتين من العازل. لقد تم تطبيق هذه الطريقة في الحقل في اسكتلندا وتم قياس شقوق وثقوب تصل إلى 5 أمتار. وقد أظهرت الدراسة أن الثقب الهيدروليكي كان معيار خاطئى لاستخدامه في دراسة انسياب هذه المواد. تعد هذه الدراسة أول دراسة تقوم باستخدام شقوق كبيرة وتعد هذه التقنية الأولى من نوعها والقادرة على تقدير سعة الثقوب بشكل افضل لاستخدامها في دراسة المخاطر الناجمة عن انسياب هذه السوائل. تعد تواصلية الشبكة خاصية أساسية في الأوساط المتشققة وان تواصلية هذه السوائل وسَعت المفهوم ليتضمن خصائص السائل أو المائع.

ABSTRACT: Chlorinated solvents are among the most widespread pollutants of groundwater. As DNAPLs (dense non-aqueous phase liquids), they can move rapidly and in complex patterns through fractures to reach and contaminate large volumes of aquifer, and then dissolve to cause significant pollution of groundwater. However, clean-up of DNAPLs in fractured rocks is virtually impossible and certainly expensive. Risk assessment should be used to decide whether the pollution is serious enough to justify major expenditure on clean-up or containment. A key aspect of risk assessment for DNAPLs in fractured aquifers is to understand how deep they are likely to have penetrated through the fracture network. This paper addresses two aspects of such predictions: measuring fracture apertures in situ and the connectivity of fracture networks with respect to DNAPLs. *Fracture aperture* is an in-situ field technique that has been developed and implemented to measure aperture variability and NAPL entry pressure in an undisturbed, water-saturated rock fracture. The field experiment also provided the opportunity to measure the wetting phase relative permeability at residual non-wetting phase saturation. The RADIO (Radial Aperture Determination by the Injection of Oil) method employs a constant rate injection of a non-toxic NAPL into a fracture isolated by a double packer array. The method was applied at the field site in Scotland, and measured apertures out to ~5m from the borehole. It showed that hydraulic aperture (from packer tests) was a poor estimator of the controlling aperture for DNAPL movement. This is the first time such large-scale aperture measurements have been made, and the technique is the first which can provide useful aperture estimates for risk analysis of DNAPL movement.*Network connectivity* is a fundamental property of the fracture system. *DNAPL connectivity* extends the concept to take account of the fluid properties.

Keywords: DNAPL, fractured aquifers, chlorinated solvents, relative permeability, fracture aperture, network connectivity.

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Why Do We Care About DNAPL in Fractured Aquifers?

Contamination of groundwater by DNAPLs, in particular the chlorinated solvents, has received much attention in recent years. Although there are many sites where significant volumes of DNAPL have been released (Huling and Weaver, 1991), their distribution in the subsurface remains uncertain. DNAPLs will spread in the subsurface, primarily under the influence of gravity, until the source term is depleted or the invading fluid is immobilised by capillary forces (Kueper and McWhorter, 1991; Mercer and Cohen, 1993). In fractured bedrock aquifers, where the DNAPL is excluded from the matrix (Ross and Lu, 1999; Wealthall, 2002), fluid movement will be limited to the interconnected fracture porosity. For many bedrock aquifers this is only 0.001-0.1% of the bulk volume of the rock mass (Mackay and Cherry, 1989). The underlying assumption of this work is that even a small amount of DNAPL would travel along way in a fractured bedrock aquifer. DNAPLs dissolve to cause significant pollution of groundwater by their component chemicals.

In the UK, the severity of groundwater pollution is determined by the risks to public health and the environment, and not by absolute concentrations. Clean-up is only undertaken if the risks justify it. For example, a DNAPL spill which only reaches shallow depths may pose risks to local receptors such as occupants of the site through vapour migration, local wells, and streams draining shallow groundwater. A spill which goes deeper will contaminate a larger volume of aquifer, and so contaminate deeper flowpaths. Pollution is then likely to present risks to more distant receptors such as public supply wells and regional groundwater discharges. Hence, it is important to understand the depth of penetration of DNAPL spills for risk assessment. Depth of penetration is controlled by the connectivity of the fracture network and fracture apertures.

Fracture Aperture

Fracture aperture is a key parameter in assessing the movement of DNAPLs. Since DNAPLs are usually non-wetting of rock relative to water, excess capillary pressure is required for them to displace the water and enter fractures; this excess is inversely proportional to the square of the aperture. Whilst fracture network properties such as spacing and orientation can be measured in the field, there are few methods for characterising fracture apertures. Hydraulic measurements of bulk permeability can be converted to hydraulic aperture (Rutqvist, 1996), but require assumptions regarding the number of fractures encountered, and are not direct measurements of physical aperture. Laboratory measurements of fracture aperture have been performed by injecting NAPLs into fractures and back calculating the fracture apertures from the entry pressure required to initiate flow (Jorgensen *et al.*, 1998; Hinsby *et al.,* 1996). The objective of this study is to describe and implement an in-situ field technique to measure aperture variability and NAPL entry pressure in a naturally occurring, water-saturated fracture. The technique is applied in a single borehole in fractured breccia and sandstone. *In situ* tests have significant advantages over laboratory tests in that the fracture aperture and infills are undisturbed, and the fracture is subject to natural compressive stress conditions which are known to influence flow characteristics (Reitsma and Kueper 1994). The new field test technique uses a controlled injection of a non-toxic NAPL into a fracture isolated by a double packer array. The test is called the RADIO (Radial Aperture Determination by the Injection of Oil) test. The resulting transient pressure signature is reverse modelled using a two-phase flow numerical model to produce an aperture profile over several metres from the borehole (Steele, 2001).

The RADIO experiment generates real two-phase flow conditions in an *in situ* fracture by injection of a NAPL into a hydraulically isolated fracture. Regulatory permission was obtained for an intentional release of up to 50L of triglyceride (as sunflower oil) directly into the aquifer. Sunflower oil is an excellent DNAPL surrogate for pressure controlled field experiments where the density of the fluid will not significantly influence the experimental results. It is a harmless light non-aqueous phase liquid (LNAPL) foodstuff with low toxicity and ecotoxicity whose degradation pathway to carbon dioxide and water is rapid and well understood. Releases of hydrocarbons directly to groundwater are strictly controlled, and this is the first time that such an experiment has been permitted to the authors' knowledge.

FIELD SITE: A research site has been established on the western edge of the Dumfries basin in south-west Scotland. The basin is of Permian age, and consists of over 1000 m of sandstones and breccias. The breccia was derived from a hardened muddy flood gravel or debris flow and has little inter-granular permeability. The Dumfries basin is a key water resource in Scotland, and produces over 50% of the local public water supply (Ball, 1997).

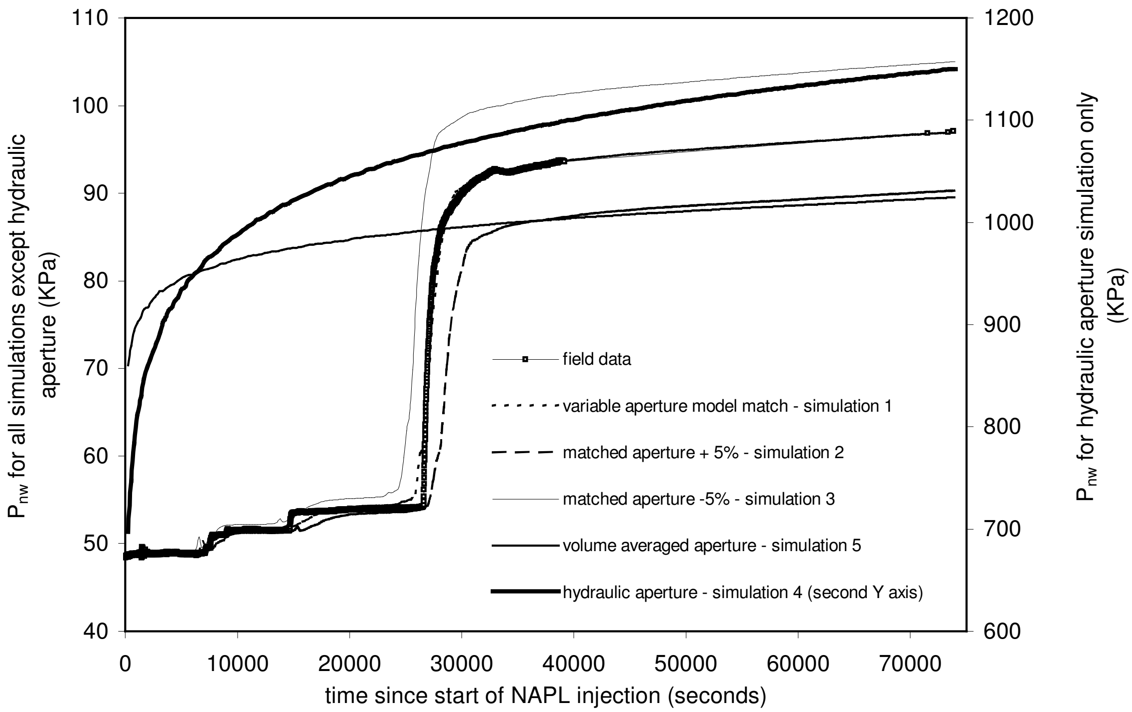

Figure 1. Transient RADIO test results and matches.

RESULTS: This experiment was the first RADIO experiment at the field site and tested the interval 13.76 m to 14.17 m below ground level which consisted only of breccia with no sandstone present. The interval hosted a single sub-horizontal fracture at 13.99 m with a dip of 19°.

The P_{nw} profile (Figure 1) shows three distinct pressure increases during the field test which lasted 73820 seconds, approximately 20.5 hours. These pressure steps were replicated in simulation 1 using the FRACAS fully compositional simulator with the resultant matched aperture profile possessing three aperture constrictions. To demonstrate the accuracy of the modeled match, the results of simulations 2 and 3 are plotted on Figure 1 with aperture profiles 5 % smaller and greater than the matched profile respectively. These runs clearly show that the resultant aperture profile presented is a comparatively accurate match to the field data.

The smallest aperture in the matched profile is 43 μm, and it is this aperture constriction that causes the large increase in P_{nw} at 27000 seconds, consequently controlling the injection pressures for most of experiment 1. Invasion of this controlling aperture is required to allow NAPL to migrate across the variable aperture fracture and we therefore denote the capillary pressure value at the leading edge of the NAPL to be the entry pressure P_e. The RADIO test therefore provides a field estimate of macro scale P_e under natural flow, stress and infill conditions. Note that 43 μm is the entry pressure aperture for distances greater than 2 m from the borehole. For distances less than 2 m, the entry pressure aperture is approximately 125 μm, illustrating clearly that the entry pressure aperture is a function of scale.

The new RADIO test can provide a much more detailed approximation of *in situ* fracture aperture than conventional hydraulic testing techniques, although it is more complicated to perform and analyse. The major constraints are the assumption of radially isotropic flow in a single horizontal fracture and the use of a literature value for S_{wr} in the numerical analysis. We have shown that aperture variations up to 5 m from the borehole can be detected and the macro scale entry pressure resolved.

Network and DNAPL Connectivity

Description of connectivity in the literature is generally limited to the physical fracture network, defined in this study as *network connectivity*. A more useful approach would be to define a connectivity expression based on the fluid active fractures, which form a sub-set of the entire fracture network. This study considers the influence of capillary pressure on the connectivity of DNAPLs in fracture networks. It therefore addresses connectivity which is both contaminant and rock specific. The connectivity study establishes a quantitative connectivity expression for DNAPL pollutants in fractured rock, which may help to define those formations that are at greatest risk from a DNAPL spill.

A probabilistic connectivity expression, C_n, is derived from multiple fracture network simulations and is based on the ratio of fractures occupied by the invading fluid to the total number of fractures in the network. The methodology has three main elements: i) simulating geometrical models by systematically varying fracture geometry, density and length in stochastically generated 3-D fracture networks; ii) calculating values of C_n; and iii) comparing C_n with published connectivity indices.

Fieldwork at the Dumfries field research site involved synthesis of existing hydrogeological data and application of multi-scale fracture characterisation methods. Quantitative field measurements of fracture parameters were obtained using 1-D scanline surveys, 2-D tracemaps (Priest and Hudson, 1981); (Gillespie *et al.,* 1993) and aperture profiling (Renshaw, 1995). Lithological and fracture logs from the rock core samples were compared with borehole televiewer logs to identify fracture type, intensity, orientation, dip and dip direction. Packered pumping tests were used to determine vertical profiles of aquifer transmissivity and calculate hydraulic aperture (Wealthall and Lerner, 2000).

RESULTS: The fracture network spatial geometry was reconstructed using a 3-D stochastic discrete fracture network model (Dershowitz *et al.,* 1988). The underlying principle of this approach is that fracture network statistics, measured at outcrop and in boreholes, can be used generate multiple realisations of fracture networks with equivalent statistical properties. Model calibration is accomplished using an inverse approach where the model realisations are sampled using simulated, yet comparable, methods to those used in the field (boreholes or traceplanes). The simulated data are then compared to the real data and adjusted till an acceptable match with the observed data is achieved. Each model realisation is just one possible representation of the fracture network from which the statistical parameters were derived. The fracture network in each realisation becomes the conductive elements for simulating fluid flow.

A fully three-dimensional, macroscopic invasion-percolation model (Wealthall, 2002) simulates the invasion of a dense non-aqueous liquid (DNAPL) in a fractured rock aquifer. The model is used to calculate bulk retention capacity and connectivity in fracture networks generated by the discrete fracture network model. Figure 2 shows an example of the results from the process, demonstrating how the DNAPL connectivity of a rock mass varies with capillary pressure. The lowest value of bulk retention capacity represents the initial entry of PCE DNAPL in the fracture network at Cr_{min}, and continues to increase until the entire fracture network that is connected to the source term is invaded at Cr_{max}.

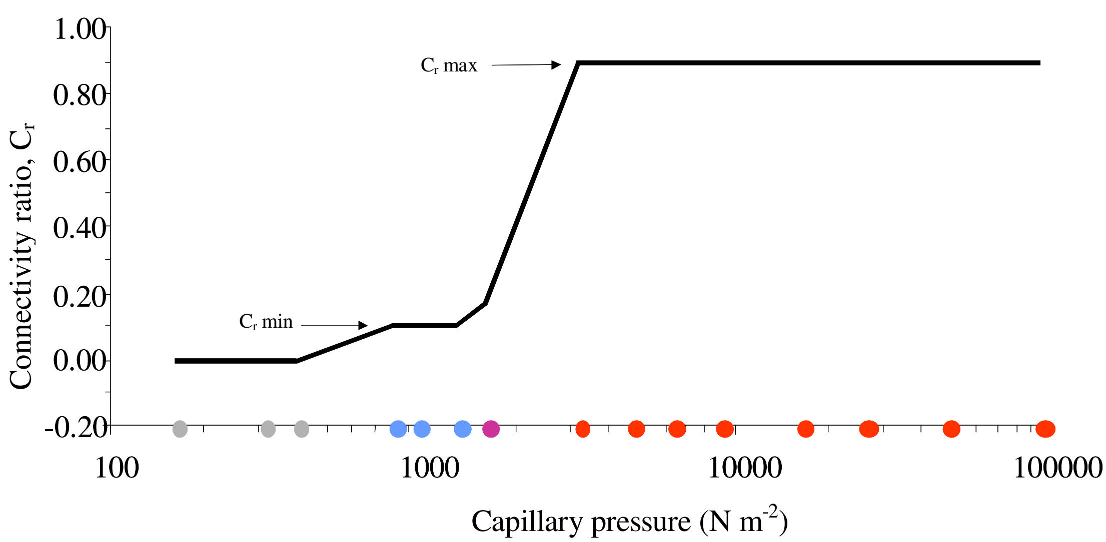

Figure 2. Principal features of the connectivity ratio plot for a single model realisation. The markers on the x-axis indicate the capillary pressure increments.

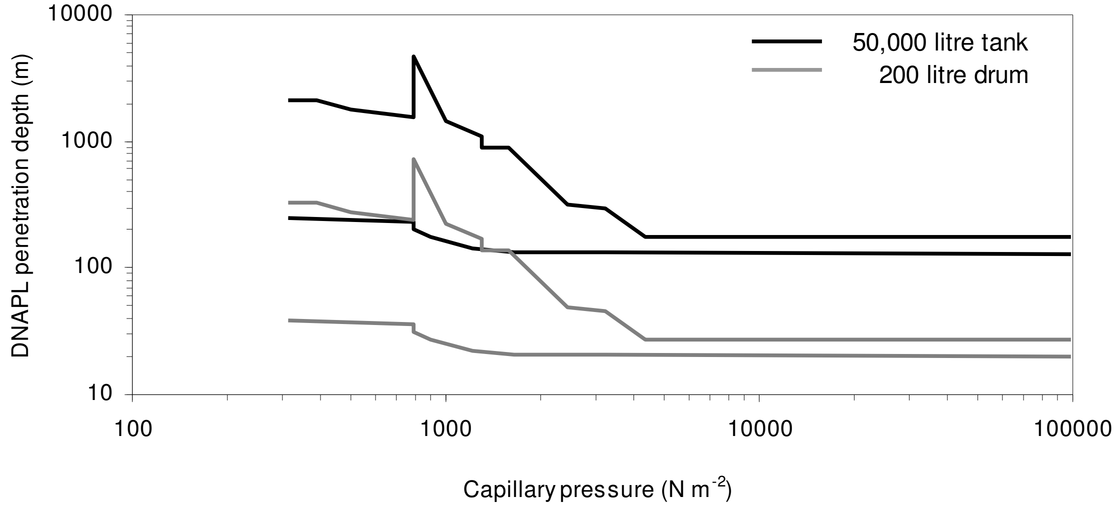

Figure 3. Effect of hypothetical spill volume on DNAPL penetration depth.

Ninety-nine models were generated with 340 fractures per realisation and, depending on individual model geometry, up to approximately 1500 fracture intersections. Cr_{min} for the bulk of the 3DIP model realisations is 800 Nm^{-1}, equivalent to a fracture aperture of 106 µm. Hypothetical spill volumes were applied to the bulk retention capacity distributions to define the depth of penetration (Figure 3). In the absence of detailed information on the geometry of the aquifer in the region of the research site, a simple cubic shaped aquifer was defined to estimate potential DNAPL penetration depth. DNAPL penetration depth is inversely proportional to capillary pressure. This reflects the low storage capacity at low capillary pressures and indicates that an given volume of DNAPL will travel much further in a low storage capacity rock mass than in a high storage capacity system. Capillary values that are in the range between the Cr_{min} and Cr_{max} may lead to high DNAPL penetration depths.

These values are summarised in Table 1 for the given hypothetical spill volumes of PCE DNAPL (outliers are not included in this table).

The fracture network in this Permian sandstone basin is well connected. A high capillary pressure release (30 cm PCE pool height) would be able to invade 90 to 100 % of the fracture network in all model simulations, whereas the low capillary pressure release (6 cm PCE pool height) would result in limited invasion of the fracture network and therefore low connectivity relative to the DNAPL.

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TABLE 1

Bulk retention capacity and DNAPL penetration depth ranges.

	Low Capillary Pressure Release	High Capillary Pressure Release
Capillary pressure ((N m^{-1})	799	3197
Equivalent PCE DNAPL pool height (m)	5	20
Bulk retention capacity (m^3 m^{-3})	8x10^{-9}	20
PCE DNAPL storage capacity (ml m^{-3})	0.008	2x10^{-5}
200 L spill: DNAPL penetration depth (m)	325	20
50000 L spill: DNAPL penetration depth (m)	2050	146

Discussion

Understanding phase occupancy in a fracture network at a given capillary pressure is critical to predicting the proportion of the fracture network that may be impacted by a DNAPL release. Given further information on residual saturation in individual fractures, an estimate of the volume of rock impacted by a finite volume DNAPL release can be inferred. A spill of 200 litres (drum volume) would penetrate to between 23 and 325 m depth, whereas a catastrophic release of 50,000 litres (tanker volume) would penetrate to between 146 and 2,050 metres depth. Both estimates assume no changes in fracture properties with depth, as this study was limited to fracture networks that were characterised in the near subsurface. Many fractured bedrock aquifer systems are developed in deep sedimentary basins, and we anticipate that connectivity and depth of penetration will vary spatially as a function of the deposition, digenesis and tectonic history of the basin. The significant findings of this study are that:

- DNAPL-effective apertures can be measured in the field, which will increase the confidence in forecasting DNAPL migration through fractured rocks.
- Uncertainty, inherent in representing the fracture network, leads to a wide range of values for bulk retention capacity and DNAPL penetration depth, particularly at low capillary pressures.
- The inferred DNAPL penetration depth is inversely correlated with capillary pressure.
- Capillary pressures, equivalent to PCE DNAPL pool heights of between 5 and 20 cm pose the greatest risk of deep DNAPL penetration to the fractured bedrock aquifer in this study.
- Comparison of connectivity measures from synthetic and real fracture networks suggests that, at least in this study, natural fracture networks in sedimentary bedrock aquifers are well connected.
- DNAPLs can be expected to penetrate deeply into fractured rocks, with lithological controls being the main barrier to downward migration. Such deep penetration leads to a high risk of polluting large volumes of groundwater, and significant risks to health and the environment.

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