Palaeo-roothole facilitated contaminant transport through clay

Science Summary SC020039-6/SS

Aromatic hydrocarbons have penetrated a clay aquitard six metres below ground surface, helped by the presence of ancient rootholes - palaeo-rootholes - dating from the time when the clay layer was exposed at the surface, according to new research commissioned by the Environment Agency. This raises concerns that the protection of underlying aquifers by such aquitards can be compromised in unfavourable palaeoenvironments.

The research was carried out by the University of Birmingham at a contaminated former industrial facility in the UK, where light non-aqueous phase liquid (LNAPL) aromatic hydrocarbons had been inadvertently released in the 1960s and 1970s. LNAPL accumulated on the shallow water table in the upper windblown-sand aquifer and gradually released dissolved-phase plumes of benzene, toluene, ethylbenzene, xylenes and styrene that are subject to ongoing investigation and remediation. The upper aquifer is underlain by a one- to two-metre thick early Holocene-age lagoonal (lacustrine) ‘intermediate clay’ aquitard at around six metres depth that was initially assumed to provide reasonable protection for the underlying marine-sand aquifer (Figure 1). Although data are sparse, some sporadic low-level contamination of the lower aquifer was found, suggesting that the aquitard may not afford complete protection of the lower aquifer. More detailed investigation was carried out to determine possible mechanisms of contaminant transport through the clay.

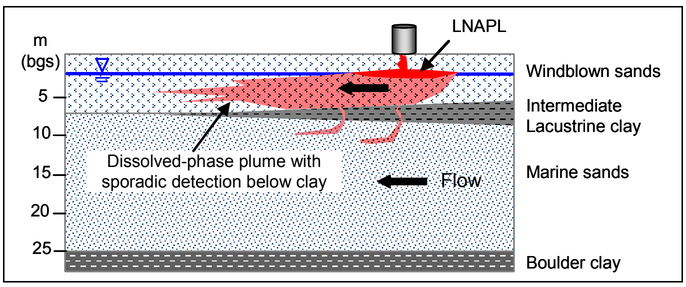

Fig. 1. Site conceptual model.

The degree of groundwater protection afforded by aquitards is an important consideration for many aquifer systems. Aquitards are often assumed to provide good protection of underlying aquifers as diffusion-controlled transport even in thin (less than two metres thick) aquitards may result in contaminant transit times of many decades or longer. However, many aquitard studies, including those on glacial tills, lake deposits, *in situ* weathered material, and floodplain deposits, have found preferential conduits which permit more rapid advective transport. Flowpaths may occur in the form of fractures, biopores (including plant root and animal hole features), discrete sand lenses and sediment-filled desiccation cracks. These units are effectively ‘dual-porosity’, comprising a porous matrix hosting a ‘pipe’ conduit network. They may lead to preferential flow and contaminant movement that are orders of magnitude greater than is possible in the fine-grain clay matrix: in the latter, flows are very low and contaminant movement typically diffusion-controlled. Previous studies on clays have usually focused on geological units at the surface. Studies of older buried aquitards like this study site are absent in the literature. Although preferential flow features will tend to close up with depth and time, until this process is complete the potential remains for more rapid advective contaminant migration. This report shows the importance of evaluating the palaeoenvironment of buried aquitards when considering contaminant migration. It is likely, for example, that sub-aerial unconformities such as the ones found here will enhance the permeability of the underlying deposits.

Thirteen cores were taken from the clay in this study. Cores from four borehole transects were analysed for volatile organic compounds (VOCs). The clay was homogeneous and unfractured, but root-like features were present in some cores. These were investigated using thin sectioning, horizontal sectioning of the cores, and micro X-ray tomography. f_{oc} (fraction of organic carbon) and K (hydraulic conductivity) characterisation, batch sorption studies and numerical modelling of contaminant transport were also undertaken.

Contaminant profiles revealed marked variation in penetration of the clay by VOCs across the site.

Profiles were shown to be either diffusion-controlled (exhibiting limited VOC penetration of the clay with exponentially declining concentrations) or advection-dominated (in which higher concentrations penetrated much further into the clay). This contrast is illustrated for benzene in Figure 2. A similar number of diffusion- and advection-type core profiles were observed. However, more core data using a gridded network approach would be required to confidently map the relative distribution of these type profiles.

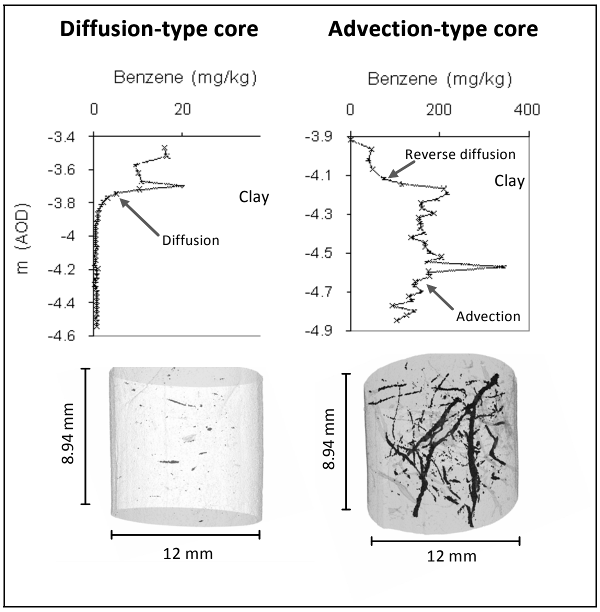

Fig. 2. Benzene profiles and X-ray tomography images.

Core inspection showed that palaeo-roothole occurrence was much greater in the cores where VOC transport was advection-dominated. Such palaeo-roothole clays were assumed to be dual-porosity in nature. From previous geological work the clay, and therefore the rootholes, were dated to around 10,000 years ago. The rootholes,, up to two mm in diameter, were well-connected in the advection-type cores but sparse and disconnected in the diffusion-type cores (Figure 2). The palaeo-roothole networks lead to bulk permeability increases of two to four orders of magnitude over the host clay, the latter having hydraulic conductivity of around 2×10^{-5} metres per day. Modelling indicates that where root networks fully penetrate the clay, advective transport is significant and may cause steady-state contaminant fluxes through the clay 10 to 1,000 times greater than that from diffusion alone. The rate of contaminant penetration is slowed by matrix diffusion and sorption which leads to similar rates of migration for each of the VOCs. More work is needed to quantify the attenuation potential and, importantly, the occurrence of biodegradation in the organic carbon-lined palaeo-roothole networks: the small pore size is likely to exclude bacteria from the clay matrix. There is some evidence that the upper part of the clay is becoming less contaminated (Figure 2) through upwards diffusion; this may be due to recent remedial works lowering concentrations in the upper sand. The variable development of rootholes is to be expected in coastal lagoonal-salt marsh type environments.

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The study has illustrated the importance of considering palaeoenvironments when assessing the likely integrity of buried clay aquitards. Palaeosol development in mudrocks, including lacustrine and estuarine clays, is generally expected to increase permeability, especially in relatively young, less indurated sequences, and hence reduce protection against migrating contaminants. In contaminated site investigation and related groundwater protection strategies, it is necessary to consider not just the main rock type present, but the aquitard's palaeoenvironment.

More generally, the study underlines the importance of using palaeoenvironmental interpretation in the development of conceptual site models. Although several of the approaches used in this study will not be viable in most contaminated land studies, recognition of palaeoenvironments is a standard geological skill. Where existing interpretations are not available or sufficiently detailed, visual inspection of exposure or core, if necessary followed by thin section and palaeontological analysis, should provide the data required for an interpretation. Qualitative implications of an interpretation will often be clear. Taking the case of a sub-aerial unconformity in a weakly indurated sequence, as studied here, if (non-calcrete) palaeosol development is significant then permeability may be enhanced to perhaps at most a few metres: palaeo-rootholes may provide greatly enhanced vertical permeability, particularly in the upper part of the palaeosol profile. Knowledge of this type is of clear use in developing conceptual models, whether they are to be used directly in decision-making, as the basis for mathematical modelling of a system, or as the basis for designing further testing programmes.

This summary relates to information from Science Project SC020039/6, reported in detail in the following:

White, R.A., 2007. Organic contaminant transport through a thin clay aquitard influenced by palaeo-heterogeneities. PhD thesis, School of Geography, Earth & Environmental Sciences, University of Birmingham.

White, R.A., Rivett, M.O., Tellam, J.H., 2008. Paleo-roothole facilitated transport of aromatic hydrocarbons through a Holocene clay bed. *Environmental Science & Technology* 42(19), 7118-7124. http://pubs.acs.org/doi/abs/10.1021/es800797u

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