



Nested monitoring approaches to delineate groundwater trichloroethene discharge to a UK lowland stream at multiple spatial scales

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ABSTRACT

Integrated approaches for the identification of pollutant linkages between aquifers and streams are of crucial importance for evaluating the environmental risks posed by industrial contaminants like trichloroethene (TCE). This study presents a systematic, multi-scale approach to characterising groundwater TCE discharge to a 'gaining' UK lowland stream receiving baseflow from a major Permo-Triassic sandstone aquifer. Beginning with a limited number of initial monitoring points, we aim to provide a 'first pass' mechanistic understanding of the plume's fate at the aquifer/stream interface using a novel combination of streambed diffusion samplers, riparian monitoring wells and drive-point mini-piezometers in a spatially nested sampling configuration. Our results indicate the potential discharge zone of the plume to extend along a stream reach of 120 m in length, delineated by a network of 60 in-situ diffusion samplers. Within this section, a 40 m long sub-reach of higher concentration ($>10 \mu\text{g L}^{-1}$) was identified; centred on a meander bend in the floodplain. 25 multi-level mini-piezometers installed to target this down-scaled reach revealed even higher TCE concentrations (20–40 $\mu\text{g L}^{-1}$), significantly above alluvial groundwater samples ($<6 \mu\text{g L}^{-1}$) from 15 riparian monitoring wells. Significant lateral and vertical spatial heterogeneity in TCE concentrations within the top 1 m of the streambed was observed with the decimetre-scale vertical resolution provided by multi-level mini-piezometers. It appears that the distribution of fine-grained material in the Holocene deposits of the riparian floodplain and below the channel is exerting significant local-scale geological controls on the location and magnitude of the TCE discharge. Large-scale in-situ biodegradation of the plume was not evident during the monitoring campaigns. However, detections of *cis*-1,2-dichloroethene and vinyl chloride in discrete sections of the sediment profile indicate that shallow (e.g., $<20 \text{ cm}$) TCE transformation may be significant at a local scale in the streambed deposits. Our findings highlight the need for efficient multi-scale monitoring strategies in geologically heterogeneous lowland stream/aquifer systems in order to more adequately quantify the risk to surface water ecological receptors posed by point-source groundwater contaminants like TCE.

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1. Introduction

Chlorinated ethenes (CEs) such as trichloroethene (TCE), tetrachloroethene (PCE) and their anaerobic metabolites

cis-1,2-dichloroethene (cDCE) and vinyl chloride (VC) are among the most problematic group of volatile organic compounds (VOCs) detected in groundwater (Basu et al., 2006; Carter et al., 2008; Rivett et al., 1990; Squillace et al., 2004). Their widespread historic use as industrial solvents has led to frequent releases to soil and groundwater associated with many individual brownfield sites (Bishop et al., 1993; Guilbeault et al.,

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2005; Witt et al., 2002) as well as large-scale contaminated industrial 'megasites' centres (Burston et al., 1993; Rivett et al., 2012; Schirmer et al., 2006). TCE has been used as a degreaser of metal parts and instances of its occurrence and persistence in aquifers are frequently documented in the literature (e.g., Rivett et al., 1990, 2005, 2012; Yang et al., 2012). As a primary solvent TCE enters the subsurface as a dense, non-aqueous phase liquid (DNAPL), capable of rapid and complex migration below the water table where it slowly migrates through the local groundwater flow system as a dissolved phase plume (Rivett et al., 2001). As a solute, TCE shows weak sorption to aquifer solids and its biodegradation is generally limited to anaerobic conditions. In aquifers with high redox potential and low sediment carbon content, migrating plumes can often be chemically unreactive and extend to distances of up to several kilometres from source areas (Benker et al., 1997; Jackson, 1998). In addition, an often close association of chlorinated ethene DNAPL sites with surface water bodies in lowland settings has been reported (Chapman et al., 2007; LaSage et al., 2008; Lorah and Olsen, 1999) including urban areas where historic industrialisation has centred on major water courses (e.g., Rivett et al., 2012). In such cases, contamination may reach shallow alluvial aquifers and intercept natural groundwater baseflow discharge to streams and rivers, with the potential to cause deterioration of surface water quality (Ellis and Rivett, 2007; Kim and Hemond, 1998; McKnight et al., 2010; Yamamoto et al., 2001).

In the European Union, adoption of the Water Framework Directive (CEC, 2000) has necessitated a more integrated approach to management of groundwater and surface water resources. Its practical implementation by member states will require an improved process-orientated understanding of the potential for pollutant exchange between these two catchment compartments in order to maintain the 'good ecological status' of dependent surface water ecosystems (McKnight et al., 2010, 2012; Smith et al., 2008). The groundwater/surface water interface of streams which comprises the saturated sediments of the channel and adjacent floodplain (also known as the hyporheic zone) is often characterised by a high degree of variability in sediment properties and hydrological exchange pathways (Allen et al., 2010; Fleckenstein et al., 2006; Huggenberger et al., 1998; Kalbus et al., 2009; Sophocleous, 2002; Woessner, 2000). As a consequence of this, there is often substantial uncertainty associated with the prediction of groundwater contaminant distribution, movement and fate in the near-stream fluvial deposits (e.g., Conant et al., 2004; Fryar et al., 2000; Milosevic et al., 2012). Within this transition zone, stratification of sediment texture, incorporation of labile organic materials and electron acceptor supply limitations may provide niche zones of enhanced microbial biogeochemical turnover where the potential for degradation of recalcitrant chlorinated organic contaminants may exceed that of the adjacent groundwater system (Abe et al., 2009; Conant et al., 2004; Hamonts et al., 2009, 2012; Schmidt et al., 2011).

Some of the first field studies investigating VOC plumes discharging to streams have involved the use of seepage meters and single-level piezometers (e.g. Norman et al., 1986). Later studies have used temperature mapping in the discharge zones of plumes to delineate areas of preferential groundwater flow through streambed deposits where a significant thermal gradient is present between surface water and underlying

groundwater (Abe et al., 2009; Conant, 2004; Milosevic et al., 2012; Schmidt et al., 2011). Temperature observations may be acquired rapidly and can potentially provide high-resolution spatial information on stream-aquifer connectivity and groundwater fluxes (Schmidt et al., 2007). However, contaminant flow lines may not always be coincident with areas of enhanced groundwater discharge where a wide range of aquifer flow paths naturally converge and additional VOC concentration data are required to validate pathway assumptions (Conant et al., 2004). Diffusion-based passive sampling approaches utilising low-density polyethylene (LDPE) membranes have gained considerable interest as potential monitoring tool in VOC-contaminated sites (ITRC, 2005). Passive sampling in streambeds offers advantages over conventional methods in that it allows the natural pore water flow regime to be maintained during sample acquisition and negates the need for purging and management of large volumes of potentially contaminated water. LDPE is a low-cost non-porous polymer that experiences transient cavitations in its molecular structure allowing small non-polar molecules to diffuse across thin (e.g., <0.1 mm) films, in response to a local concentration gradient (Vrana et al., 2005) and is known to be compatible with all the CEs (Ehlike et al., 2004; Vroblesky and Campbell, 2001). Passive vapour samplers, utilising partitioning of pore water VOCs to the air space of vial enveloped by an LDPE membrane have been used to delineate VOC plumes impacting benthic sediments (Church et al., 2002; Lyford et al., 1999; Vroblesky and Hyde, 1997; Vroblesky and Robertson, 1996; Vroblesky et al., 1991, 1996). The approach is cost effective as a screening tool for reconnaissance surveys in that it provides for a high density of in-situ monitoring points to be quickly established in plan-view, without the prerequisite of existing infrastructure such as monitoring wells.

However, in order to provide an integrated assessment of contaminant fate at groundwater/surface water interfaces, vertical profiling at sediment-scale (e.g., 0.1 to 1 m) is necessary to capture physical and chemical gradients which may exert significant local controls on the transport and degradation potential of VOC plumes. For example, Lorah and Olsen (1999) used transects of nested piezometers to evaluate redox conditions and in-situ degradation of multiple chlorinated VOCs (including TCE) along upwelling groundwater flow paths through reducing sediments of a tidal wetland. This is one of the few field studies to demonstrate the potential for efficient natural attenuation of chlorinated organic compounds at groundwater/surface water interfaces. Similarly, Conant et al. (2004) used the 'waterloo profiler' (Pitkin et al., 1999), multi-level sampler bundles and drive-point piezometers to investigate the potential discharge of a shallow PCE plume in riverbed and riparian sediments in Ontario, Canada. The high vertical resolution of their sampling approach revealed intricate details of plume heterogeneity with concentrations changes of an order of magnitude within a spatial scale of 1 m. The authors were able to demonstrate substantial transformation of the PCE plume within the riverbed deposits by detection of the PCE degradation products (mainly cDCE and VC) in vertical pore water profiles. Similarly, in the UK, Roche et al. (2008) used drive-point mini-piezometers (Rivett et al., 2008) to demonstrate partial dechlorination of a TCE plume discharging to a 50 m instrumented reach of the highly urbanised River Tame in Birmingham, UK (Ellis and Rivett,

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