Journal of Contaminant Hydrology 171 (2014) 1–11

Contents lists available at ScienceDirect

Journal of Contaminant Hydrology

journal homepage: www.elsevier.com/locate/jconhyd

High-resolution delineation of chlorinated volatile organic compounds in a dipping, fractured mudstone: Depth- and strata-dependent spatial variability from rock-core sampling

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ARTICLE INFO

Article history:
Received 11 September 2013
Received in revised form 29 July 2014
Accepted 6 October 2014
Available online 12 October 2014

Keywords:
Trichloroethene
Dichloroethene
Vinyl chloride
Lockatong Formation
Newark Basin
Bedding-plane fractures
Monitored natural attenuation (MNA)
DNAPL
Site conceptual model
Methanol extraction

ABSTRACT

Synthesis of rock-core sampling and chlorinated volatile organic compound (CVOC) analysis at five coreholes, with hydraulic and water-quality monitoring and a detailed hydrogeologic framework, was used to characterize the fine-scale distribution of CVOCs in dipping, fractured mudstones of the Lockatong Formation of Triassic age, of the Newark Basin in West Trenton, New Jersey. From these results, a refined conceptual model for more than 55 years of migration of CVOCs and depth- and strata-dependent rock-matrix contamination was developed. Industrial use of trichloroethene (TCE) at the former Naval Air Warfare Center (NAWC) from 1953 to 1995 resulted in dense non-aqueous phase liquid (DNAPL) TCE and dissolved TCE and related breakdown products, including other CVOCs, in underlying mudstones. Shallow highly weathered and fractured strata overlie unweathered, gently dipping, fractured strata that become progressively less fractured with depth. The unweathered lithology includes black highly fractured (fissile) carbon-rich strata, gray mildly fractured thinly layered (laminated) strata, and light-gray weakly fractured massive strata. CVOC concentrations in water samples pumped from the shallow weathered and highly fractured strata remain elevated near residual DNAPL TCE, but dilution by uncontaminated recharge, and other natural and engineered attenuation processes, have substantially reduced concentrations along flow paths removed from sources and residual DNAPL. CVOCs also were detected in most rock-core samples in source areas in shallow wells. In many locations, lower aqueous concentrations, compared to rock core concentrations, suggest that CVOCs are presently back-diffusing from the rock matrix. Below the weathered and highly fractured strata, and to depths of at least 50 meters (m), groundwater flow and contaminant transport is primarily in bedding-plane-oriented fractures in thin fissile high-carbon strata, and in fractured, laminated strata of the gently dipping mudstones. Despite more than 18 years of pump and treat (P&T) remediation, and natural attenuation processes, CVOC concentrations in aqueous samples pumped from these deeper strata remain elevated in isolated intervals. DNAPL was detected in one borehole during coring at a depth of 27 m. In contrast to core samples from the weathered zone, concentrations in core samples from deeper unweathered and unfractured strata are typically below detection. However, high CVOC concentrations were found in isolated samples from fissile black carbon-rich strata and fractured gray laminated strata. Aqueous-phase concentrations were correspondingly high in samples pumped from these strata via short-interval wells or packer-isolated zones in long boreholes. A refined conceptual site model considers that prior to P&T remediation groundwater flow was primarily subhorizontal in the higher-permeability near surface strata, and the bulk of contaminant mass was shallow. CVOCs diffused into these fractured and weathered mudstones. DNAPL and high concentrations of CVOCs migrated slowly down in deeper unweathered strata, primarily along isolated dipping bedding-plane fractures. After P&T began in 1995, using wells open to both shallow and deep strata,

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http://dx.doi.org/10.1016/j.jconhyd.2014.10.005
0169-7722/Published by Elsevier B.V.

downward transport of dissolved CVOCs accelerated. Diffusion of TCE and other CVOCs from deeper fractures penetrated only a few centimeters into the unweathered rock matrix, likely due to sorption of CVOCs on rock organic carbon. Remediation in the deep, unweathered strata may benefit from the relatively limited migration of CVOCs into the rock matrix. Synthesis of rock core sampling from closely spaced boreholes with geophysical logging and hydraulic testing improves understanding of the controls on CVOC delineation and informs remediation design and monitoring.

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1. Introduction

Remediation of groundwater at fractured-rock sites contaminated with Dense Non-Aqueous Phase Liquids (DNAPL) and dissolved chlorinated volatile organic compounds (CVOC) remains a major challenge. A recent strategic study by the U.S.'s National Research Council (NRC) (2013) noted "The most problematic [hazardous waste] sites are those with potentially persistent contaminants including chlorinated solvents recalcitrant to biodegradation, and with hydrogeologic conditions characterized by large spatial heterogeneity or the presence of fractures." In fractured-rock settings, especially sedimentary rocks, the rock matrix is a reservoir for contaminants that can act as a long-term secondary source by DNAPL dissolution, back diffusion, and de-sorption (Dearden et al., 2013; Mutch et al., 1993; Parker et al., 2010; Révész et al., 2014).

Characterization of the CVOC mass distribution in fractured sedimentary rocks has recently advanced using rock core sampling techniques, providing a fine scale vertical profile of CVOC mass diffused into or sorbed onto rock strata (Sterling et al., 2005; Parker et al., 2010). Analysis of rock core for CVOCs provides fine-scale delineation of contaminants in and on the rock matrix, and is not affected by possible vertical contaminant migration in the borehole after coring (cross contamination). Dearden et al. (2013) used rock core sampling to identify the CVOC profile into about 4 m of Triassic-age mudstone underlying a surficial aquifer. Chapman et al. (2013) recently presented rock core sampling results from 3 coreholes in fractured sedimentary rocks of the Newark Basin, with depths of about 70, 75, and 120 m, respectively, and horizontal spacings of more than 200 m apart along a flow path.

Our study extends previous interpretation of the rock core sampling results by synthesis with borehole geophysical methods and multi-level water-quality monitoring within a site conceptual model based on a detailed hydrogeologic framework, using closely spaced coreholes of depths of up to 60 m. This approach allows a site-scale characterization of the control of weathering and strata-bound permeability features on CVOC migration and persistence in sedimentary fractured rocks. Our study also provides additional direct confirmation of some of the fine scale features included in current conceptual models of CVOC migration in fractured sedimentary rocks, such as limited diffusion into deep unweathered rock. These results are used to refine the conceptual model for CVOC fate and transport in fractured sedimentary rock aquifers.

1.1. Site description

In this investigation, we synthesize closely spaced rock core sampling results with multi-level water-quality monitoring, borehole geophysical logging, and hydraulic testing at the former Naval Air Warfare Center (NAWC), West Trenton, New Jersey (Fig. 1). Lacombe (2000, 2002, 2011) describes monitoring well configurations at the site and interpretation of water levels and contaminant concentrations measured in monitoring wells installed by the U.S. Navy. Industrial use of TCE at the NAWC resulted in DNAPL TCE, and dissolved TCE, cis-1,2-dichloroethene (DCE), and vinyl chloride (VC) in groundwater, and historic contaminant discharges to Gold Run, a culverted stream along the site's southern boundary (Fig. 1). The distribution of peak CVOC concentration reflects source areas where DNAPL TCE was released, and is still present, and migration of dissolved CVOCs towards Gold Run. Since the mid-1990s a pump and treat (P&T) system has minimized discharge of CVOCs beyond the site boundary and to Gold Run (Lewis-Brown and Rice, 2002).

The evolving conceptual site model for contaminant migration at NAWC is based on a detailed hydrogeologic framework (Lacombe, 2000; Lacombe and Burton, 2010). The study area is underlain by mudstones of the Lockatong Formation of Triassic age, of the Newark Basin, one of a series of early Mesozoic Basins in the eastern U.S. in which mudstone, shale, and sandstone aquifers are used for water supply (Trapp and Horn, 1997). At NAWC the mudstone strata are gently dipping to the NW, and a fault along the southeastern boundary of the site separates the Lockatong from the older sandstone of the Stockton Formation (Fig. 1). The detailed lithology of the local mudstones is divided into distinctly colored strata that occur in cycles of deposition: thin (less than 0.2-m thick) black strata containing up to 7% organic carbon by weight (Marjorie S. Schulz, USGS, written comm., 2007; Lebron et al., 2013) and generally highly fractured (fissile), gray mildly fractured thinly layered (laminated) strata, and light-gray weakly fractured massive strata. Several of the high-carbon fissile black beds are herein designated by their respective depths in corehole 43BR at the site: for example the top of a black fissile stratum designated "BlkFis-233" occurs at a depth of 233 ft (71 m) in 43BR.

Below relatively high-permeability weathered and highly fractured strata, groundwater flow is primarily in bedding-plane fractures and thin fissile high-carbon strata, or in fractured, laminated strata in the dipping mudstones, as identified by borehole (Williams et al., 2007) and aquifer testing (Lacombe, 2000; Tiedeman et al., 2010). Large-scale horizontal anisotropy in transmissivity is a well-known characteristic of Newark Basin aquifers, with preferential water-level responses to pumping in the direction of strike of the dipping beds (Herpers and Barksdale, 1951; Longwill and Wood, 1965; Vecchioli, 1965). This apparent horizontal anisotropy is caused by the dipping hydro-stratigraphy, together with the vertical anisotropy of hydraulic conductivity (HK): HK is highest parallel to the dipping beds and lowest perpendicular to beds (Michalski, 1990; Michalski and Britton, 1997; Goode and Senior, 1998, 2000; Senior and Goode, 1999; Lacombe, 2000).

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