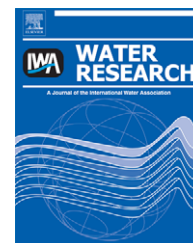


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Influence of organic carbon loading, sediment associated metal oxide content and sediment grain size distributions upon *Cryptosporidium parvum* removal during riverbank filtration operations, Sonoma County, CA

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ABSTRACT

This study assessed the efficacy for removing *Cryptosporidium parvum* oocysts of poorly sorted, Fe- and Al-rich, subsurface sediments collected from 0.9 to 4.9 and 1.7–13.9 m below land surface at an operating riverbank filtration (RBF) site (Russian River, Sonoma County, CA). Both formaldehyde-killed oocysts and oocyst-sized (3 μm) microspheres were employed in sediment-packed flow-through and static columns. The degree of surface coverage of metal oxides on sediment grain surfaces correlated strongly with the degrees of oocyst and microsphere removals. In contrast, average grain size (D_{50}) was not a good indicator of either microsphere or oocyst removal, suggesting that the primary mechanism of immobilization within these sediments is sorptive filtration rather than physical straining. A low specific UV absorbance (SUVA) for organic matter isolated from the Russian River, suggested that the modest concentration of the SUVA component (0.8 mg L^{-1}) of the 2.2 mg L^{-1} dissolved organic carbon (DOC) is relatively unreactive. Nevertheless, an amendment of 2.2 mg L^{-1} of isolated river DOC to column sediments resulted in up to a 35.7% decrease in sorption of oocysts and (or) oocyst-sized microspheres. Amendments (3.2 μM) of the anionic surfactant, sodium dodecyl benzene sulfonate (SDBS) also caused substantive decreases (up to 31.9 times) in colloid filtration. Although the grain-surface metal oxides were found to have a high colloid-removal capacity, our study suggested that any major changes within the watershed that would result in long-term alterations in either the quantity and (or) the character of the river's DOC could alter the effectiveness of pathogen removal during RBF operations.

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1. Introduction

Riverbank filtration (RBF) is a widespread water management operation where bank sediments are used as a pre-treatment option for substantively reducing the quantity of

many common microbial and chemical contaminants (Tufenkji et al., 2002). The Enhanced Surface Water Treatment Rule (LT2ESWTR) of the U.S. Environmental Protection Agency (EPA, 2006) specifies criteria for pathogen removal and grants treatment credits to utilities employing bank

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filtration as part of their overall drinking water treatment process. Because of episodic high abundances of oocysts of the protozoan pathogen *Cryptosporidium parvum* in surface waters (Rose, 1997; M. LeChevallier et al., 1999, M.W. LeChevallier et al., 1999), their resistance to chemical disinfection (Macler and Merkle, 2000; Coffey et al., 2007), and their longevity in the environment (Ives et al., 2007), their removal is a key parameter for assessing the efficacy of RBF sites in the United States. Models have been applied to *C. parvum* oocyst transport through porous media (Harter et al., 2000; Darnault et al., 2004), but there is little detailed information about transport of oocysts between the riverbed and water supply wells. A recent study by Weiss et al. (2005) concluded that simple monitoring for pathogens – which can occur episodically at RBF sites, may be inadequate for assessing log removal rates (and therefore treatment credits). They suggested that further studies, including column tests, are necessary in order to assess the transport character and removal rates for pathogen surrogates and indicator organisms during passage through RBF systems. Finally, Tufenkji (2007) concluded that more accurate and complete models of microbe fate and mobility in porous media would require well-designed laboratory column experimentation in assessing microbial transport.

The subsurface fate and transport of *C. parvum* oocysts depend on a variety of physical, biological and geochemical parameters (Harvey et al., 2007), including flow rate, temperature, pH, dissolved organic compounds (Dai and Hozalski, 2002), ionic strength (Tufenkji et al., 2004), sediment grain size distribution, metal oxide content (Abudalo et al., 2005), microbe size, and both microbe and sediment surface charge (Tufenkji et al., 2004). In one of the few in-situ studies assessing *C. parvum* oocyst transport at operating RBF sites, Metge et al. (2007) reported that oocyst-sized microspheres were readily removed within the first meter of travel in both riverbed and infiltration basin sediments. However, removal of oocyst-sized colloids within deeper, underlying sediments had not been studied.

This paper presents results and assessments from laboratory studies designed to provide information regarding the roles of the salient aspects of the sediment and river chemistry on the transport of oocyst-sized colloids at the Russian River RBF site. The overarching goal of this study was to determine the contributions of total extractable metals, grain-surface metal oxide coverage, grain size, and the character and amount of DOC upon colloidal removal within the subsurface sediments sampled between 1 and 14 m below land surface (bls) near operating RBF wells. A secondary objective involved assessment of the spatial variability in filtration efficiency of *C. parvum* oocysts within the Russian River sediments along vertical transects. In addition to the use of killed oocysts, we also employed carboxylated microspheres, which were used in pilot-scale filtration systems to assess removal efficiency for oocysts (Emelko and Huck, 2004) and in the aforementioned field study (Metge et al., 2007). Because the surface characteristics of oocyst-sized microspheres are subject to change to a lesser extent than oocysts, microspheres can be valuable surrogates that facilitate comparisons among different transport studies.

2. Materials and methods

2.1. Site characterization

A brief overview of the Russian River watershed and study site is found in Metge et al. (2007) and Anders et al. (2006). With flow varying seasonally, the Russian River watershed (area = 3850 km²) extends 160 km from its headwaters in the Coast Range in Mendocino County, California, to the Pacific Ocean near Jenner, California (Rantz and Thompson, 1967). A U.S. Geological Survey (USGS) gauging station near Guerneville measures mean daily flows, which vary seasonally from 4.5 to 265 m³ s⁻¹ (<http://nwis.waterdata.usgs.gov/ca/nwis/>, site no. 11467000).

The Sonoma County Water Agency (SCWA) operates a bank filtration system of 6 wells along the lower reach of the Russian River in Northern California near Forestville, California. The wells collectively have a maximum production capacity of over 14,500 m³ h⁻¹ (92 million gallons day⁻¹) with another 3200 m³ h⁻¹ (20 mgd) standby capacity (Su et al., 2007). These facilities utilize natural filtration processes to provide water for about 600,000 people in Sonoma and Marin Counties. The wells extract water from the unconsolidated alluvial aquifer adjacent to and beneath the Russian River via large-volume Ranney-type (lateral) collector wells. The laterals are located about 25 m below the bottom of the river. The pumping wells induce large vertical fluxes from the river and nearby infiltration ponds. During the summer months, an inflatable dam is erected to create an enhanced river stage and establish a large hydraulic gradient between the river and collector wells.

Measured hydraulic conductivities for the shallow aquifer sediments site range from 5.5×10^{-5} to 2.0×10^{-4} m s⁻¹ (Su et al., 2004) and from 1.4×10^{-5} to 2.6×10^{-4} m s⁻¹ within the same area using seepage meter techniques (Gorman, 2004). Three slug tests conducted in shallow sampling wells indicated a moderately good connection between surface water and ground water with hydraulic conductivities ranging between 2×10^{-5} m s⁻¹ in fine-grained medium sands in recharge basins to 8×10^{-5} m s⁻¹ in coarser sand or sand and gravels within the river. River and groundwater within the system are slightly alkaline (pH 7.3–8.5 in the river and pH 7.5–8.5 in infiltration ponds) (Metge et al., 2007). The river water has modest electrical conductivity levels (214–598 μ S cm⁻¹) and ionic strength of ~ 3 mM. Porosity ranged from 23% (Russian River sediments) to 30% (infiltration ponds).

2.2. Sediment size fraction and mineralogical analyses

We collected sediment cores, using split-spoon recovery techniques (Fetter, 1988), from two locations (well 1 at 38°30'41" N, 122°52'57" W and well 2 at 38°30'41" N, 122°53'00" W) near operational RBF wells operated by the Sonoma County Water Agency (SCWA) as shown in Fig. 1A. The cores were shipped to the USGS laboratory in Boulder, CO. The depth intervals used in our study were between 1.7 and 13.7 m taken at 1.5-m intervals and between 0.9 and 4.9 m at 0.3 m-intervals. Core materials also were obtained near the infiltration ponds (38°29'49" N, 122°53'28" W) as shown in Fig. 1A.

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