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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.

1. Introduction

Riverbank filtration (RBF) is a widespread water management operation where bank sediments are used as a pretreatment 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 km^2) 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 $m^3\ s^{-1}$ (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 $m^3\ h^{-1}$ (92 million gallons day^{-1}) with another 3200 $m^3\ h^{-1}$ (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 $\mu S\ cm^{-1}$) 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.

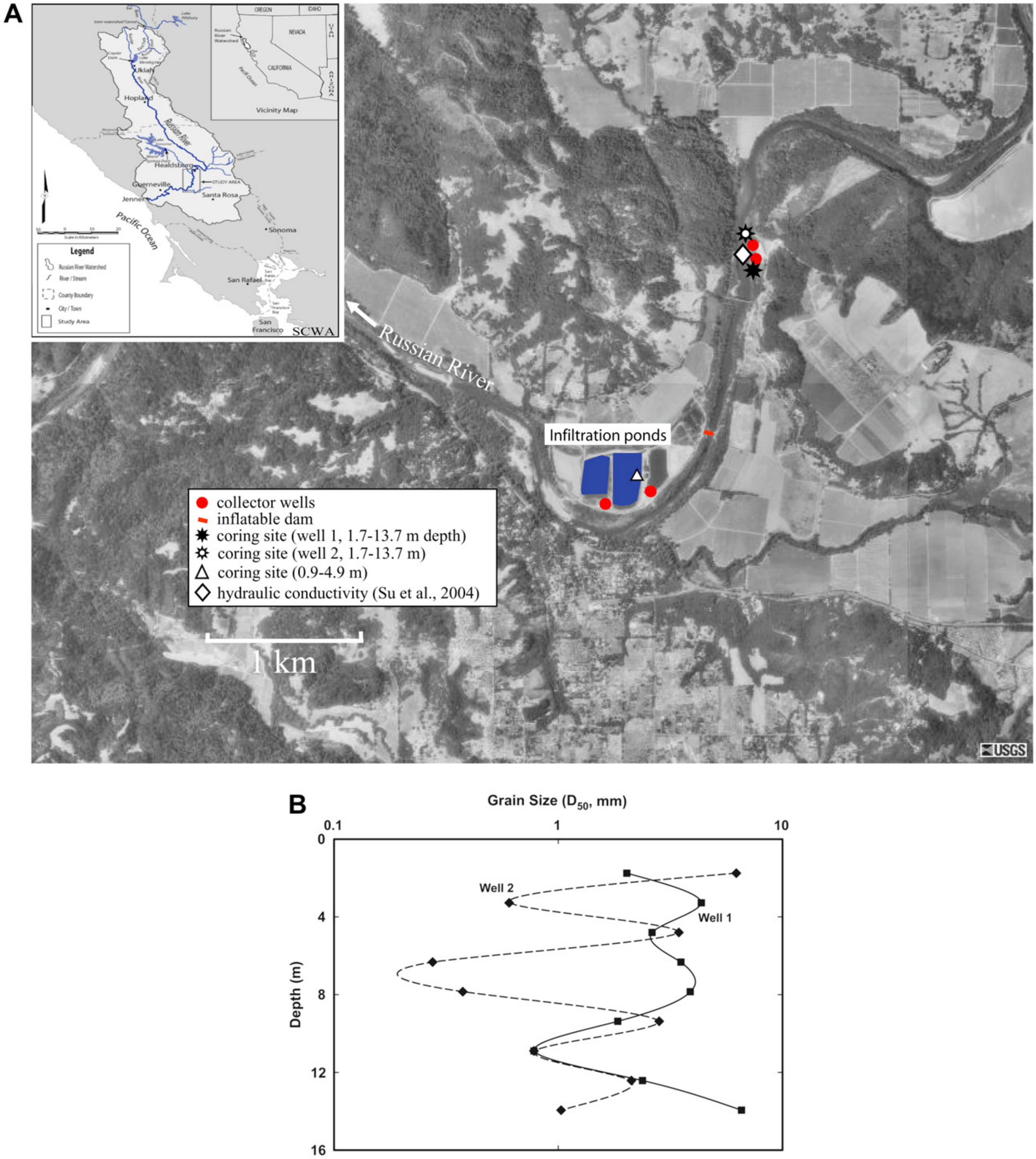

Fig. 1 – A – Location of coring sites. Relative locations of Well 1 and Well 2 split-spoon coring sites, intermediate depth split-spoon coring sites, collector wells, inflatable dam, and approximate location of Su et al. (2004) hydraulic conductivity study. B – Average grain size plotted as a function of depth from two locations (Well 1, ■, and Well 2, ◆) near production wells at Sonoma County Water Agency's Russian River bank filtration site.

Subsurface sediments from each depth interval were dry sieved with sieve sizes ranging from <0.063 to 16 mm. We determined the D_5 to D_{95} sizes (including D_{50} values) for each sediment interval. D values are the weight-normalized percentages of sediment grains passing through screens of specific mesh sizes. The D_{10}-values were used for calculating hydraulic conductivities (K) using the Hazen equation (Odong, 2008). These estimations were compared with estimates from previous aquifer tests (Su et al., 2004; Gorman, 2004).

Extractable metals (EPA method 3050) were assessed at the University of Colorado, Boulder LEGS (Laboratory for Environmental and Geological Studies) facility. All metal results were normalized to total dry sediment weight and to the specific size fractions in order to obtain metal distributions for bulk and sieved sediment fractions. Scanning electron microscopy (SEM) analyses were performed using a JEOL 5800LV electron microscope (JEOL Ltd, Tokyo, Japan) equipped with an Oxford ISIS (EDX, energy dispersive X-ray) and Noran NSS analyses system (ThermoFisher Scientific, Madison, WI). Sample preparation and SEM procedures are detailed in Lowers et al. (2005a, 2005b). Sediment samples were carbon-sputtered before assessment of grain mineralogies according to methods outlined in Lowers et al. (2005a). We also determined if oocysts and fluorescent microspheres were associated with particular mineralogies by EDX analyses of grain areas in where colloids were proximal to the sediment grain(s). Acceleration voltages and magnifications were 20.0 kV and 1200–6000×, respectively.

2.3. *Oocysts and microspheres*

An HS-Bryte (Biorad Corp., Hercules, CA) 3-color flow cytometer with Apogee 1.3 software (Apogee, London, UK) and an Hg excitation lamp (OSRAM, HBO 103 W/2) were used to enumerate Polysciences YG type microspheres (excitation and emission wavelengths approximately 486 nm and 521 nm, respectively) and fluorescently labeled *C. parvum* oocysts (DNA-specific fluorescent dye 4,6-diamidino-2-phenylindole, DAPI, 0.1 mg L^{-1}, final concentration, 30–90 min staining time). DAPI has excitation and emission spectra at 359 and 461 nm, respectively. BG1 and GR1 (Blue-green-1 and Green-1, Biorad, Hercules, CA) filter cubes separate the specific emission spectra and pass signals to respective photomultiplier tubes. The polydispersed colloids were differentially quantified on the basis of both size and fluorescence intensity. Each sample was assayed in triplicate for up to 300 s. The coefficient of variation (CV) for flow cytometry varied from 3.8 to 7.6% for 3.9×10^4 to 4.0×10^6 microspheres (2 μm-diameter) mL^{-1} and from 11.1 to 19.0% for 3.5×10^2 microspheres mL^{-1} to 3.8×10^3 microspheres mL^{-1}. Coefficients of variation for fluorescently-stained *C. parvum* oocysts ranged from 6.1 to 14.8% for abundances in the range of 3.2×10^5 mL^{-1} to 8.5×10^5 mL^{-1} and from 21.0 to 39.2% for lower abundances of 3.0×10^2 to 3.0×10^4 oocysts mL^{-1}.

We obtained formalin-inactivated *C. parvum* oocysts from Sterling Parasitology Laboratory (SPL), University of Arizona, Tucson. The oocysts were harvested from a calf infected with the "Iowa" isolate of *C. parvum* (Dr. Harvey Moon, National Animal Disease Center, Ames, Iowa) and purified at the SPL before shipment to the USGS, (Boulder, CO) as described in Abudalo et al. (2005) and Harvey et al. (2008). Before the laboratory study, oocysts were pelleted as previously described (Harvey et al., 2008), stained with the DNA-specific fluorescent dye DAPI (0.1 mg L^{-1}, final concentration, 30–90 min staining time), filtered through a 1.0-μm pore size filter, washed with 1 mM NaCl to remove excess stain and any bacteria in suspension, and resuspended in 1 mM NaCl. The final oocyst concentration was 10^5 to 10^7 oocyst mL^{-1}. The peak excitation and emission wavelengths for DAPI-stained oocysts are 359 and 461 nm, respectively. Oocyst abundances within the range of 1.3×10^6 to 5.8×10^1 oocysts mL^{-1} had an associated coefficient of variation ranging from 4.4 to 28.9%, respectively.

2.4. *Organic carbon*

A large-volume water sample was collected from the Russian River and shipped on ice via overnight courier to the USGS laboratory in Boulder, CO. Upon arrival, the sample was filtered with Gelman AquaPrep 600 capsule filters (0.45 μm) that were prerinsed with 2 L of sample water. Dissolved organic carbon (DOC) concentration was determined using an OI Analytical 700 TOC analyzer (Aiken, 1992). UV–visible absorbance measurements were performed on a Hewlett Packard photo-diode array spectrophotometer (model 8453) using a 10 mm quartz cell. Specific UV absorbance (SUVA254) was determined by dividing the UV absorbance measured at 254 nm by the DOC concentration and reported in units of milligrams carbon per liter according to methods outlined in Weishaar et al. (2003). Concentrations of aquatic humic substances were measured chromatographically by following the procedures described by Aiken (1992).

2.5. *Flow-through column experiments*

Column experiments were conducted as detailed Abudalo et al. (2005). Sodium nitrate ($NaNO_3$, 1 mM) was used as a conservative tracer and added as a pulse via a 5 mL injection loop. Individual fractions were collected using a fraction collector (Eldex Corporation, Napa, CA) and measured by ultraviolet absorption (220-nm wavelength) using a spectrophotometer (Spectronic/Unicam, Genesys 10, Thermo Scientific, Pittsburgh, PA). The transport experiments were conducted at room temperature (25 °C) within glass chromatography columns (2.5 × 10.0 cm length, Spectrum Chromatography, St Louis, MO) with polytetra-fluoroethene end caps and polypropylene mesh (105-μm pore size) on both column ends. Columns were dry-packed with milli-Q (18 Ω–Ohm) washed bank filtration sediments from specific intervals and purged with 2% CO_2 gas. Approximately 10 pore volumes of degassed background solution (1 mM NaCl, pH 5.6–5.8) were passed through the column at a flow rate of ∼2.9 m d^{-1} flow rate using a digital piston pump (stainless steel, 500 mL volume, Isco 500D, Omaha, NE) and polypropylene tubing. Effluent pH and specific conductance of inlet and outlet solutions were monitored to ensure pH stability before introducing the conservative tracer and colloids.

Filtered (0.45-μm pore size, cellulose acetate filters) 1 mM NaCl was amended with isolated river water DOC (final concentration of 2.2 mg DOC L^{-1}) and introduced as 5 mL pulses to flow-through columns of subsurface sediments. This was done to condition the sediments prior to addition of oocysts or microspheres. Effluent DOC levels were measured after ∼4 pore volumes had passed through each column. Tracer solutions and colloid (microsphere and oocyst) suspensions were added to the background solution using a high performance liquid chromatography injector (stainless steel; Supelco Rheodyne, Sigma Aldrich, St. Louis, MO) and injection loop (stainless steel, 5.0-mL volume for microspheres and oocysts). A fraction collector and glass test tubes were used to collect the column effluent.

Total particle load was maintained at approximately 1.3×10^7 total particles. Effluent fractions were assayed by flow cytometry as described above and in Metge et al. (2007). Oocyst and microsphere recoveries were normalized and compared to pore volume. Column porosities in all flow-through experiments ranged from 29.1 to 34.6%. These estimated porosities were determined gravimetrically and the estimated values were confirmed by analyses of the tracer breakthroughs. Attachment was estimated from the number of colloids recovered and plotted as a function of average grain size and extractable metals to assess the relative effect of either physical or chemical sorption upon transport through these sediments.

2.6. *Static mini-column experiments*

The sorption-desorption studies with oocysts and microspheres were conducted with 20 mL static mini columns (2.5-cm diameter, 12-cm height) modified from that described by Scholl and Harvey (1992). Acid-washed and baked (450 °C) glass columns were prepared with polypropylene mesh (105-μm pore size) as support for sediments. Ten mL of 1 mM NaCl solution (pH 5.6–5.8) was added to approximately 10 g of SCWA sediment. All experiments were conducted at room temperature (22–25 °C) and in triplicate.Columns were equilibrated by rinsing with at least 20 pore volumes of background solution. One pore volume (PV) was ~2–2.5 mL when using static columns with SCWA sediments. The oocyst-microsphere suspension (1 mL of 1.37×10^7 total particles) was carefully added to the top of the sediment in each column and drawn into the sediments by withdrawing fluid from the bottom of the column. The columns were subsequently equilibrated for 4 h, after which the pore fluid was collected in acid washed glass vials. A 20- PV "chaser" volume of sterile background solution was subsequently passed through the column in order to remove and collect any unattached microspheres or oocysts. The abundances of recovered oocysts and microspheres were quantified by flow cytometry or, if the eluent was turbid, by epifluorescence microscopy as described in Section 2.3. Fractional attachment of the colloids was determined from the difference between the number added and number recovered from each column.

The effect of grain size and sediment surface-associated metal contents upon microsphere or oocyst attachment was assessed by placing different sediments with different grain sizes and extractable metal contents within the static column. An experiment with SDBS (3.2 μM, 1 mg L^{-1} SDBS- sodium dodecyl benzene sulfonate) was performed in triplicate with three sediment types (low, medium and high metal oxide contents). For comparison, a 1 mM NaCl solution (pH 5.6) was added to replicate columns in order to determine the effect of SDBS-free systems upon microsphere or oocyst attachment.

3. Results

3.1. *Sediment characterization*

Fig. 1B illustrates the disparate manner in which average grain size (D_{50}) varies with depth for the coring sites near wells 1 and 2. These coring sites were shown in Fig. 1A. Despite differences in average grain size with depth between the two locations, there was a high degree of uniformity with respect to extractable metals contained on collected sediments as summarized in Table 1. Iron (Fe) comprised a majority (~62%) of extractable metals (EPA method 3050). Aluminum (Al) contributed ~37% whereas manganese (Mn) comprised only ~1%. These proportions appeared to be relatively constant by depth and location. Iron and aluminum are the dominant (~97%) extractable metal species present on sediment surfaces. The presence of metals, particularly Fe, Mn, and Al, on grain surfaces was confirmed by scanning electron microscopy (SEM)-EDX (energy-dispersive X-ray spectroscopy) microanalyses of sediment grains from different strata within the coring profiles. A typical scan is illustrated in Fig. 2. These grains contained areas of elevated Fe and Al which are the lighter or shinier region(s) labeled photo insert and visible on the left photo. From the images, it appeared that microspheres, which are negatively charged at the pH of the experiments, tended to associate with patches of iron-based minerals, based upon EDX point analyses (right photo, Fig. 2).

Grain-size distributions (D_{10} values) from the intermediate (0.9–4.9 m) and deeper (1.7–13.7 m) depth intervals were used to calculate hydraulic conductivities by the Hazen relationship. Hydraulic conductivity (K) is a measure of the ease by which water is capable of moving through porous media, and is function of grain size distributions. Calculated K values were plotted as a function of extractable iron (Fig. 3) from the sediments. Results indicate that higher metal content was associated with larger grain sediments in samples taken from intermediate depths (0.9–4.9 m). By contrast, data from a deeper depth interval (1.7–13.7 m) produced an inverse relationship – i.e., extractable metal contents decreased with increasing estimated hydraulic conductivities.

Table 1 – Distribution of iron (Fe), aluminum (Al), and manganese (Mn) in Russian River Sediments used in SDBS experiment (Fig. 5). Analyses by EPA method 3050 were performed at, LEGS Laboratory at the CU, Boulder campus. Lower half of table shows relative contributions of each metal type to total metal and was based upon metal distributions for Well 1 and 2 cores.

Sediment type (metal content)	Fe (mg g^{-1} sediment)	Al (mg g^{-1} sediment)	Mn (mg g^{-1} sediment)	Total Metals (mg g^{-1} sediment)
Low	19.75	11.47	0.30	31.52
Medium	23.40	14.84	0.30	38.55
High	34.29	19.94	0.49	54.72
Relative contribution	Fe % of total	Al % of total	Mn % of total	
Mean ± SE	61.9 ± 0.43	36.6 ± 0.45	1.5 ± 0.19	
Range	60.7–69.7	29.9–38.7	0.6–4.4	

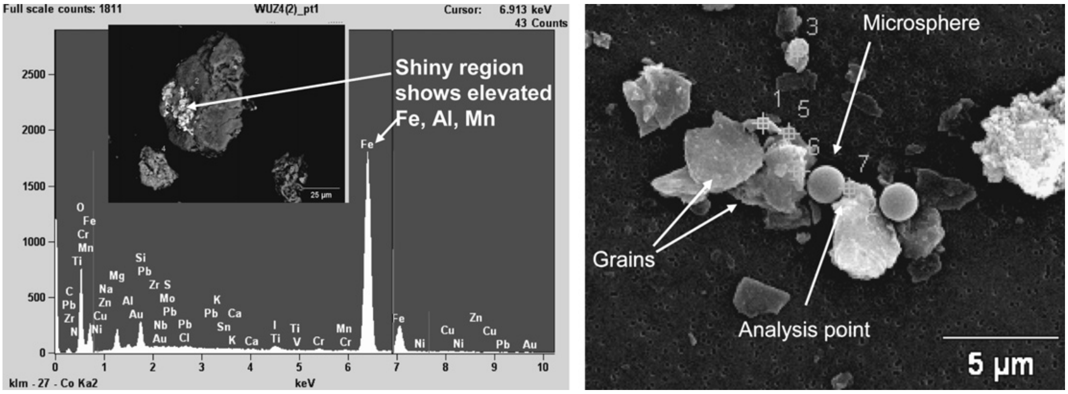

Fig. 2 – Scanning electron microscopy of Russian River sediment surfaces. Left picture shows EDX scan of shiny area in photo insert and indicating area of high Fe, Mn, and Al. Numbers on photo insert indicate points that were scanned as well. Histogram in left picture shows relative amount of specific elements and an example of grain area with higher metal oxide content. Right photograph is an SEM picture of 2-μm microsphere with sediment grains. Numbers indicate EDX measurement points.

3.2. *Effect of extractable metals and grain size on colloid transport in RBF sediments*

A series of static experiments were performed in order to assess how grain size (Fig. 1B) and sediment metal content (Table 1) affected degree of colloid transport Normalized oocyst and microsphere attachment did not correlate with grain size (D_{50}) values. Correlation coefficients (R^2) ranged from 0.19 to 0.27 ($p < 0.08$–0.12) indicating that attachment was not significantly related to grain size. However, there was a very strong first order relationship between the quantity of total extractable metals in the sediments and degree of removal for the 3-μm (diameter) microspheres ($R^2 = 0.69$, $p < 0.025$) and between total extractable metals and removal of oocysts (and $R^2 = 0.82$, $p < 0.01$) as shown in Fig. 4. Significant first order correlations ($p < 0.01$–0.05) were evident between total extractable metals and removal of 2- and 5-μm sized microspheres. The first-order relationships were observed for both static mini-columns and flow-through column experimental systems.

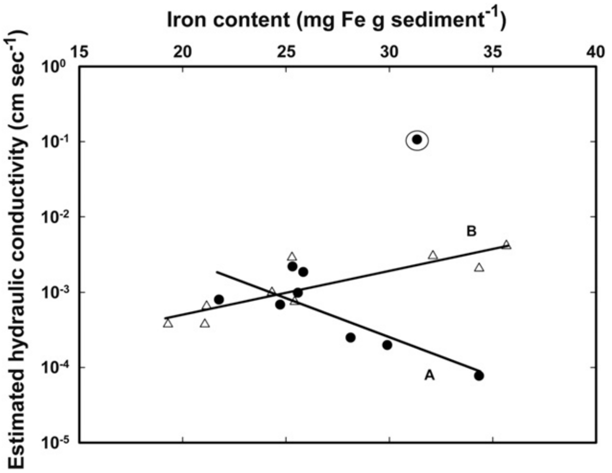

Fig. 3 – RBF sediment hydraulic conductivities for selected depth intervals plotted as a function of Fe-content. Based upon grain size distribution analyses, hydraulic conductivities (K) were estimated using the Hazen equation. These K values were plotted as a function of extractable iron (Fe) values normalized by sediment weights. Two relationships were determined. Closed circles (●) are data from sediments over the entire Well 1 depth profile (A). Open triangles (△) are data from sediments at intermediate depths (B) located below infiltration ponds. Data point in dashed circle was considered an outlier and not included in regression analyses. Curve A (Well 1 sediments from 1.5 to 13.7 m): $y = 0.43e^{-0.25x}$, $R^2 = 0.66$ and Curve B (Intermediate depth sediments 0.9–4.6 m): $y = 6 \times 10^{-5}e^{0.12x}$, $R^2 = 0.72$.

3.3. *Effect of surfactant and naturally occurring organic material (NOM) on colloid transport in RBF sediments*

The effects of modest additions (1 mg L^{-1}, final concentration) of the anionic surfactant SDBS upon attachment of microspheres and oocysts are shown in Fig. 5 (left and right graphs, respectively). Table 1 shows the relative distribution of Fe, Al and Mn in the three sediment types used in the experiment. For all sediments tested, the presence of SDBS decreased attachment or removal of microspheres and oocysts. The effect of SDBS upon microsphere and oocyst attachment increased linearly with extractable metal content such that the greatest effect of SDBS upon colloid transport occurred in the presence of sediments having the highest extractable metal content. The effect of natural organic matter (NOM) isolated from Russian River water on microsphere and oocysts removal was assessed (Fig. 6). NOM (2.2 mg L^{-1}) was used to pretreat sediments (containing 20.1 mg Fe g^{-1} sediment) within flow-through columns. Attachment of oocysts decreased slightly, (86.0–90.2%), after NOM addition, whereas microsphere attachment decreased modestly (2.9 μm FMS, 77.5%–65.0%) to substantially (4.9 μm FMS, 96.7%–61.0%).

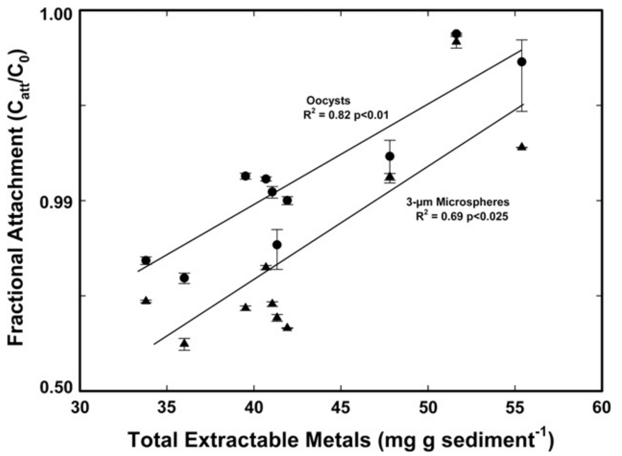

Fig. 4 – Effect of sediment extractable metal content upon oocyst and oocyst-sized (3-μm) microsphere removal from static columns. Fractional removal from static column experiments for oocysts (●) and 3-μm diameter microspheres (▲) plotted as a function of total extractable metals for corresponding sediments. Error bars are standard deviations of the mean from replicate columns for each data point. The relationship between 3-μm removal and metal content was: $y = 1 - 14{,}356e^{-0.30x}$, $R^2 = 0.69$ and the relationship between oocyst removal and metal content was $y = 1 - 407.18e^{-0.26x}$, $R^2 = 0.82$. The Y-axis is a probit scale from 0.50 to 0.9999 (rounded to 1.00). This reflects calculated removal levels of oocysts and microspheres within these column systems.

4. Discussion

4.1. *Sediment hydraulic, chemical and mineralogical properties*

It is well-established that hydraulic conductivity (K) is related to grain-size distributions of porous media (Freeze and Cherry, 1979). However, in the Russian River RBF system, it was unclear how or if K would be related to extractable iron and aluminum oxides that affect the sorptive filtration of microspheres or oocysts (Fig. 4). A variety of equations have been developed which allow hydraulic conductivity calculations using grain size analyses, D-values (fraction of sample passing through a mesh screen of specific size), and porosity which are presented in Odong (2008). Although such calculations are only estimates, our estimates of K (3.4×10^{-4} to 8.8×10^{-6} m s^{-1}) were consistent with values reported for the Russian River site in previous studies, e.g., from 5.5×10^{-5} to 2.0×10^{-4} m s^{-1} (Su et al., 2004), 4.17×10^{-4} m s^{-1} (Constantz et al., 2006) and 1.4×10^{-5} to 2.6×10^{-4} m s^{-1} (Gorman, 2004).

An assessment was made as whether there was a relationship between K calculated using the Hazen equation (Odong, 2008), and sediment-associated metals. Although, in the deeper interval sediments, there was a significant ($R^2 = 0.66$, $p < 0.01$) but inverse relationship between K and sediment-associated iron; the intermediate-depth sediments appeared to have a direct relationship between sediment metal content and K (Fig. 3, $R^2 = 0.72$, $p < 0.01$). These results suggested that grain surface-associated metals were more likely associated with different (larger or smaller) grains depending upon depth interval or site sampled. Surprisingly, there were no significant linear relationships between K and microsphere or oocyst attachment as R^2 values were low ($R^2 = 0.07$–0.14, $p = 0.30$–0.14, respectively) for Well 1 and 2 sediments. Consequently, hydraulic conductivity variations may be of limited value in predicting variability of pathogen removal at this particular site.

4.2. *Influence of sediment grain size and grain-surface chemistry*

Fig. 1B illustrates the average grain-size distributions over two depth profiles within bank filtration sediments near two operational bank filtration production wells. The sediments consist largely of quartz sand, with minor amounts of clays. Table 1 illustrates the relative concentration and contribution of Fe, Al and Mn to the extractable metal content range typically found within these sediments. Despite rather substantive differences in grain size, distributions in extractable metal oxides appear similar throughout all sediments collected from the SCWA's bank filtration site. Previous work (Metge et al., 2007) suggested that because removal rates for oocysts closely traced those of microspheres over several orders of magnitude and because grains were somewhat rounded, factors controlling attachment behavior of the two colloids likely involved the same mechanism(s). For straining to occur in fairly uniform to mixed model quartz grains, Bradford et al. (2002), suggested that a colloid to average grain diameter (D_p/D_g) of >0.0017 was necessary. In subsequent research using quartz sands, Bradford and Bettahar (2005) and Bradford et al. (2003, 2004; 2005) found that the aforementioned ratio for straining should be >0.003–0.005. However, Tufenkji et al. (2004) suggested a ratio of >0.018 for angular (but model) grains in order for straining of oocysts to occur. Tufenkji's research suggested that mean grain to particle size cannot be used as a sole predictor of straining potential but indicated that straining is likely an important factor in natural heterogeneous systems. Additionally, Tufenkji and Elimelech (2005) point out that laboratory experiments with model sediments cannot account for the various factors, namely metal oxide coatings and organic matter that impact the degree of oocyst removal. These factors will enhance oocyst deposition (iron oxide coatings) (Elimelech et al., 2000) or perhaps increase transport as in the case of organic matter (Dai and Hozalski, 2002).

For the different subsurface sediments employed in our study, none of the D_p/D_g ratios were greater than 0.018, but ~35% did fall between 0.005 and 0.017. Furthermore, the aquifer sediment grains in our study were somewhat rounded with some angularities, suggesting that straining at this site likely occurs when Dp/Dg ratios are considerably higher than 0.017. Consequently, straining would not appear to be a major mechanism for immobilization of oocysts and microspheres within the deeper sediments of the Russian River RBF site, but may deserve further study or attention. This is despite the fact that an order of magnitude difference exists in D_p/D_g ratios suggested as straining controls and this study found reduced microsphere and oocyst removal efficiencies when RBF

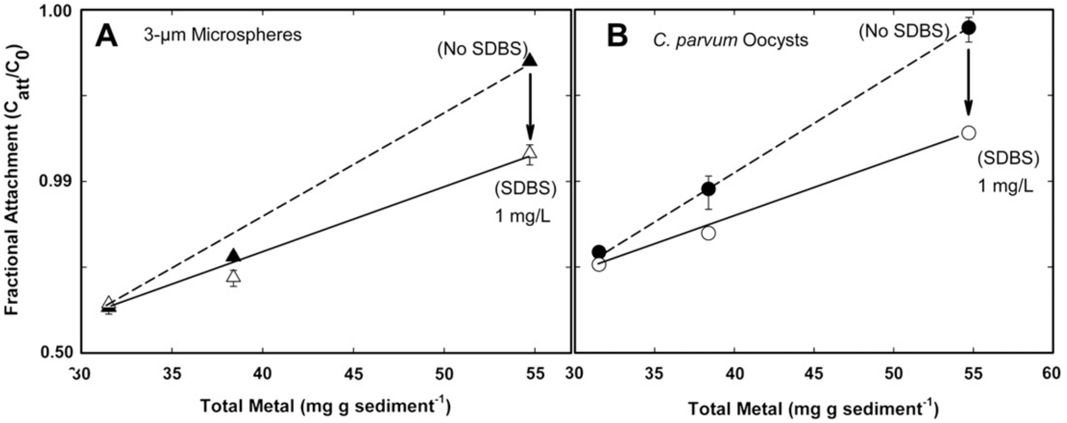

Fig. 5 – Effect of SDBS surfactant content upon oocyst-sized microsphere and oocysts attachment in static columns. Fractional removal from static column experiments for 3-µm diameter microspheres (▲) and oocysts (●) during passage through SDBS-treated (open symbols) or untreated (closed symbols) static column systems. Error bars are standard deviations of the mean from replicate columns for each data point. The Y-axis is a probit scale from 0.50 to 0.9999 (rounded to 1.00). This reflects calculated removal levels of oocysts and microspheres within these column systems.

sediments were pretreated with NOM or SDBS surfactant. No significant relationships were found between colloid removal and grain size-related parameters (i.e., grain size, estimated hydraulic conductivity or K) underscoring the conclusion by Tufenkji et al. (2004) that *Dp*/*Dg* ratios are of limited value in predicting straining.

Removal of microspheres and oocysts was very sensitive to the quantity of extractable metals (Fig. 4). An approximately 2-log unit difference in the degree of removal was accompanied by less than a two-fold change in grain surface-associated metal content. Both the 3-µm diameter microspheres and oocyst attachment displayed a log-linear relationship with metal oxides over at least two orders of magnitude and we conclude that the mechanisms of removal for both are possibly due to sorption via attractive forces between the positively charged metal oxides and negatively-charged oocysts. This type of relationship was also observed for oocysts and all microsphere classes within both laboratory flow-through columns and static mini-column methods for the various core materials examined ($N = 29$ column experiments). Other research has shown that sediment-associated metal oxide content, particularly the surface coverages of the metal oxides on the grain surfaces, is responsible for controlling virus (Pieper et al., 1997) and oocyst transport (Abudalo et al., 2005) in iron-laden quartz sand. Tufenkji et al. (2006) suggested that 3.5–3.8-µm diameter oocysts should experience a deep attractive secondary energy minimum as they approach a grain surface. Although not measured directly, the lack of tailing in breakthrough curves suggests that the reversibility of oocysts and microsphere attachment within the Russian River site sediments was low, at least under experimental conditions.

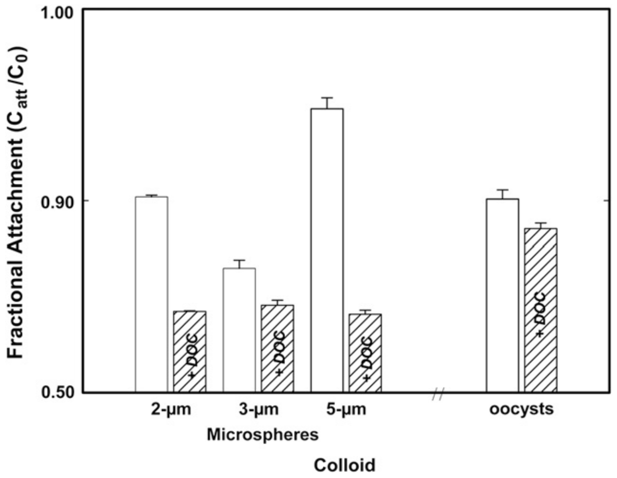

Fig. 6 – Effect of organic-free (clear bars) and organic-amended waters (hatched bars) upon oocysts and oocyst-sized microsphere fractional attachment within flow-through columns. Error bars represent propagated errors from estimated standard errors within each fraction collected. Sediment metal oxide content was 20.1 mg Fe g sediment^{-1}. The Y-axis is a probit scale from 0.50 to 0.9999 (rounded to 1.00). This reflects calculated removal levels of oocysts and microspheres within these column systems.

4.3. *Role of surfactants and NOM*

Results (Fig. 5) indicate that very modest additions (1 mg L^{-1}) of SDBS surfactant can decrease the sorptive capacity of aquifer sediments for oocysts and microspheres up to 30-fold. Anionic surfactants have been previously used in field studies to examine their effect upon microbial transport (Pieper et al., 1997). Pieper et al. (1997) reported that 87% of previously sorbed bacteriophage (PRD-1) was remobilized in the presence of 25 mg L^{-1} linear alkylbenzene sulfonate (LAS). Under acidic conditions, the amorphous iron oxyhydroxides that characterize the Cape Cod aquifer sediments for the field test (Ryan et al., 1999) exhibit a strong net positive charge. Changes in the net charge caused by a sorbing anionic surfactant should lead to a decreased propensity of negatively charged colloids for attachment. In the present study, which was also run under moderate ionic strength (10^{-3} M) and slightly acidic conditions (pH 5.6–5.8), the range of iron oxyhydroxide content examined were comparable to that reported for the Cape Cod site (∼3–5%). Both oocysts and microspheres have a negative surface charge at experimental pH (Metge et al., 2007).

SDBS is used in a variety of health care products such as shampoos and dish soaps and, consequently, can often be found in surface waters that are impacted by human activity, particularly ones that receive treated or untreated sewage effluent. If incomplete degradation of anionic surfactants during waste water treatment (Cserháti et al., 2002; Mungray and Kumar, 2008) occurred, future wastewater discharges into the Russian River would likely increase levels of anthropogenic organic compounds within water being drawn through the sediments beneath the river to the lateral collector wells. Although the effect of SDBS upon transport of pathogens in granular aquifers is not well understood, it appears that actions which change the content and character of organic matter (including compounds of anthropogenic origin) within the Russian River could affect removal rates of oocysts within these RBF sediments.

Flow-through column experiments (2.9 m day^{-1}) employing untreated and NOM-amended sediments suggest that the dissolved organic matter in the Russian River substantively enhanced transport of microspheres and colloids (Fig. 6). In the absence of NOM additions, microsphere and oocyst transport within low iron oxide sediments (20.1 mg Fe g^{-1} sediment) was modest. In columns with sediments pretreated with natural organic matter (2.2 mg C L^{-1}), a two to seven-fold increase in microsphere transport and a substantive (∼35.7%) increase in oocyst transport resulted (Fig. 6). This occurred despite our observations that the NOM is relatively unreactive, judging from the SUVA values of approximately 2.0. The retardation factor for the colloids became less than unity, suggesting enhancement of colloidal transport velocity most likely due to size-exclusion from at least a portion of the finer porosity. It has been reported that dissolved organic carbon (DOC) enhances electrostatic repulsions between the oocysts of *C. parvum* and glass beads which led to decreased attachment (Dai and Hozalski, 2002). Another study by Franchi and O'Melia (2003) used humic acid and microspheres; they observed that the negatively charged humic acid (1 mg C L^{-1}) adsorbed to both the colloid (latex microspheres) and the silica collectors. Franchi and O'Melia reported this resulted in additional electrostatic and steric contributions to the repulsive energy barrier for attachment and enhanced the reentrainment of deposited particles in porous media. Thus, the presence of humic substances results in a greater propensity for transport at low ionic strengths and may be similar to our observations that modest applications of NOM increased the degree of oocyst and microsphere transport within flow-through columns containing bank filtration sediments. These findings further underscore the fact that the RBF sediment removal efficiency for pathogens may be significantly altered or affected by both the character and quantity of DOC (anthropogenic or NOM) character.

4.4. *Use of microspheres as surrogates for oocysts*

Largely for reasons of safety, microspheres have been used as surrogates in studies involving transport and attachment of oocysts in granular media (Emelko and Huck, 2004). Also, it is generally easier and more cost effective to enumerate microspheres than oocysts (Amburgey et al., 2005). In previous field studies at the Russian River RBF site, fluorescent, carboxylated, polystyrene microspheres (Polysciences, Warrington, PA and Invitrogen-Molecular Probes, Carlsbad, CA) were used to assess the potential of the near-surface sediments to filter oocysts (Metge et al., 2007). This approach, using surrogates and oocysts introduced to columns containing sediments from intermediate (0.9–4.9 m) to deeper (1.7–13.7 m) intervals, provided additional comparisons with results from our previous field work. Finally, microspheres, evaluated as surrogates for oocysts in filtration and transport experiments, provided conservative estimates of oocyst removal in water treatment filters (Dai and Hozalski, 2003). This latter study also concluded that microsphere transport should be greater than or mimic oocyst transport – depending upon system ionic strength and organic carbon concentrations.

Carboxylated polystyrene microspheres, as abiotic surrogates for microorganisms, have been used to examine oocyst removal in water filtration systems (Emelko and Huck, 2004). Recent field-scale injection and recovery studies successfully employed carboxylated microspheres as surrogates for oocysts to investigate the transport potential of oocysts in the vicinity of municipal wells that draw water from karst (Harvey et al., 2008) and granular aquifers (Metge et al., 2007). Bacteria-sized microspheres (from 0.2 to 1.0 μm-diameter sizes) are considered relatively poor analogs in predicting bacterial transport within porous granular media (Harvey et al., 1989). However, it has been reported that microspheres within size classes bracketing groundwater protists and oocysts (2–5-μm-diameter) display transport behaviors similar to those of groundwater nanoflagellates (protozoa) (Harvey et al., 1995).

Microorganisms and microspheres differ in terms of motility, growth and surface structure. Recent research recognized that these differences are mitigated by similarities in surface charge, sorption-desorption control dynamics, and remobilization potential (Johnson et al., 2007). Another study using glass-bead columns showed that 4.1-μm carboxylated microspheres exhibited transport behavior similar to that of *C. parvum* oocysts (Tufenkji et al., 2004). More recently, similar removal efficiency for oocysts and microspheres were reported for a small-scale filtration system (Amburgey et al., 2005).

For field and laboratory applications, the utility of microspheres as surrogates for *C. parvum* oocysts depends on how accurately microspheres represent oocyst removal and transport properties. Recent studies were undertaken with microspheres as complements or surrogates to investigate microbial transport under a variety of geochemical and hydrologic conditions. These are outlined in Amburgey et al. (2005) and Johnson et al. (2007). In a pilot-scale assessment of water treatment filter removal efficiencies, Amburgey et al. (2005) found log removal rates for oocysts and microsphere surrogates were similar during conventional or biological filter tests in cold weather (11 °C) and warm weather (24 °C) conditions, however, they did find significant removal differences between oocysts and microspheres during warm weather runs with suboptimal coagulant conditions. Our study used commercially available, fluorescent, carboxylated, polystyrene microspheres as surrogates for oocysts in conjunction with formalin-killed *C. parvum* oocysts for static and flow-through column experiments with sediments collected from the SCWA's Russian River site. In addition, we performed all experiments at 22–25 °C and under consistent conditions except as noted (e.g., NOM or SDBS treatments). Although carboxylated microspheres may be obtained that have similar buoyant densities, sizes, and aspect ratios to those of oocysts (Harvey et al., 2008), the electrophoretic mobilities can differ, depending upon the pH, age, and treatment of the oocysts (Brush et al., 1998; Butkus et al., 2003). All oocysts used in these experiments were prepared in a consistent manner (Harvey et al., 2008).

5. Summary and conclusions

1. Although earlier field studies at the Russian River RBF site (Metge et al., 2007) indicated that the fine, near-surface sediments are characterized by a high capacity for immobilization of oocyst-sized colloids, the present studies indicate that the deeper, coarser sediments at the site also exhibit a high efficacy for removal of oocyst-sized colloids.
2. The efficient removal of microspheres and oocysts in the deeper sediments was clearly a function of the iron and aluminum oxide content on the grain surfaces. Removal by straining did not appear to be significant in the deeper sediments and neither average grain size nor grain-size distribution were good indicators of either microsphere or oocyst removal.
3. The presence of 1 mg L^{-1} (3.2 μM) of the model anionic surfactant, SDBS, had a substantive effect in facilitating transport of the oocysts and microspheres through the metal oxide-laden subsurface sediments, similar to findings from the Cape Cod site. More surprising was the observation that low concentrations of relatively unreactive Russian River NOM also exhibited this same effect.
4. Although the 25 m of poorly-sorted, high-iron and –aluminum sediments between the river bottom and lateral collector wells offer a natural protective barrier against transport of oocysts, the efficacy of this barrier could be affected by any future long-term changes in either the amount or the character of the dissolved organic material (natural or anthropogenic) within the Russian River.

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