Virus removal efficiency of ceramic water filters: effects of bentonite turbidity
Cameron Farrow, Edward McBean and Hamidreza Salsali

ABSTRACT
Ceramic water filters (CWFs) are utilized in many developing countries as point-of-use (POU) water treatment devices, to reduce waterborne pathogens in potable water. Virus removal efficiencies of several CWFs are investigated under various influent conditions using MS2 (ATCC: 15597-B1) as a surrogate phage for human enteric viruses. The addition of bentonite turbidity (6–8 NTU) in the influent source water showed increased viral removal efficiency of CWFs by 0.1–0.2 log compared to tests involving clear (<1 NTU) influents. Trials employing an applied clay cake layer, formed using highly turbid influent source water (100 NTU) and no cleaning regime between trials, resulted in viral removal efficiency values of 1.5–2.5 log, compared to 0.2–0.5 log during non-obstructed trials.

Key words | bentonite clay, ceramic pot filter (CPF) enteric virus, ceramic water filter (CWF), diarrheal disease, drinking water, filtration, MS2 coliphage, point-of-use (POU) water treatment

INTRODUCTION
Safe drinking water is defined as water which does not present significant health risks over a lifetime of consumption (WHO 2011). The World Health Organization stipulates that access to safe drinking water represents a basic human right, but an assessment by UNICEF found that 1.1 billion people do not have access to safe drinking water (also termed ‘improved drinking water sources’) (UNICEF 2000). In response to this challenge, many organizations have been implementing point-of-use (POU) drinking water treatment technologies in developing countries where access to centralized water supply systems is limited and/or not available.

To put the magnitude of the shortfall in provision of safe water into perspective, an estimated country-specific 66% of Cambodian residences do not have access to improved drinking water supplies (Brown 2007). To make matters worse, an even greater proportion does not have access to microbiologically safe water at the POU due to inadequacies in water storage.

A promising POU treatment technology that is capable of attaining substantial improvements with regard to microbial water quality is the ceramic water filter (CWF) (van Halem 2006; Brown 2007; Murphy et al. 2010), a POU water treatment system currently used in Nicaragua, Honduras, Cambodia and many other developing countries (Figure 1).

CWFs are constructed primarily of clay and fine/milled rice husk. Rice husk is added prior to firing to increase porosity and ensure adequate flow rates (1.5–3.0 L/h) when the CWFs are used as a POU water treatment technology. Many recipes for CWFs include laterite, a soil type rich in iron oxide, to aid in the removal of bacteria and heavy metals. The ability of POU treatment systems to remove Escherichia coli, as well as heavy metals and other contaminants has been assessed (Clasen et al. 2004; van Halem 2006; Brown 2007; Murphy et al. 2010; Murphy 2010). An alternative POU water treatment system, the Katadyn Ceradyn, consisting of a ceramic filter element, has met the EPA Guide Standard for Microbial Purifiers with regard to bacteria (99.9999% reduction) and protozoa (99.9% reduction) removal (Clasen et al. 2004). However, the ability of POU water treatment systems to efficiently remove viruses has...
not been thoroughly evaluated and yet viruses pose very significant challenges for the provision of safe water.

All viruses require a host cell for replication (Levy et al. 1994). Therefore, viruses which infect humans will not reproduce in the external environment. In contrast, some bacterial species may multiply in water supplies (WHO 2014). Due to the low infectious dose for viruses in comparison to most bacteria (WHO 2014), the degree of filtration required to reduce viral contamination below infectious doses is greater than the contamination reduction required for most bacterial species. In addition, bacteria are filtered from source water more efficiently by CWFs. When evaluating specific pathogens, factors such as persistence in the environment, capability of multiplying in water supplies, and severity of illness after infection must be assessed. For example, *Campylobacter* has a low infectious dose in comparison with other bacteria and results in severe illness upon infection. Therefore, *Campylobacter* presents similar health concerns and removal requirements to viral contaminants.

Potters for Peace, a non-profit organization that originally began manufacturing CWFs in Nicaragua, stipulates that the maximum pore size of their CWFs is 1 μm. However, more thorough examination of the filter elements by surface electron microscope (SEM) revealed that non-uniformities present within the CWF result in cracks up to 150 μm and the pore size distribution ranged between 0.6 and 5 μm (Lantagne 2001). van Halem (2006) also investigated pore size of CWFs and found the effective pore diameter to be between 33 and 52 μm using bubble point tests.

Bacteria range in length from 1–50 μm; however, rod-shaped bacteria (including *E. coli*) are 0.3–1.5 μm in diameter and 1–10 μm in length (Tchobanoglous et al. 2003). The majority of *E. coli* species have lengths at the lower end of the indicated range. Human enteric viruses range in size from 0.02–0.08 μm (Tchobanoglous et al. 2003), 100–1,000 times smaller than most bacteria. As a result, the mechanisms that govern virus removal in CWFs are expected to be very different from the methods of bacterial removal. Neither viruses nor bacteria are expected to be removed during CWF operation by size exclusion alone.

Coliphages (viruses which infect bacteria) were employed as surrogates for human enteric viruses for a variety of reasons, including: ease of detection, low cost of assay, and previous evidence indicating coliphages have a greater resistance to environmental factors (e.g. disinfection) compared to human enteric viruses (Tchobanoglous et al. 2003). MS2, the coliphage utilized in the study reported herein, is smaller than most viruses and therefore may be less efficiently removed by CWFs in comparison to other viruses.

MS2 has an isoelectric point of 3.9 and therefore carries a surface charge at neutral pH, which affects sorption properties. Most viruses have isoelectric points in the range of 3.5–7 (Michen & Graule 2010), and therefore are moderately comparable to MS2 with regards to surface charge. Viruses with higher isoelectric points will carry a lower surface charge at neutral pH, be less likely to sorb to other particles and, therefore, may be less efficiently removed, in comparison to MS2, during CWF operation. Data regarding isoelectric points of viruses are inconsistent within a single virus species and therefore difficult to use as an accurate predictor of sorption processes (Michen & Graule 2010).

Viruses that create exposure hazards to humans via consumption of contaminated water are primarily those that infect the gastrointestinal tract (enteric viruses) and thus are excreted in the faeces of infected humans (WHO 2011). These include the Hepatitis A virus, Noroviruses and Rotavirus. Human faecal matter may contain more than 100 different viruses, at concentrations in the range of 10-10,000 viruses per gram of faeces.
10^6 viruses/mL of faecal matter (Tchobanoglous et al. 2003). Raw wastewater typically contains between 10^2 and 10^5 enteric viruses/mL (Tchobanoglous et al. 2003). Given that the typical infectious dose for viruses is 1–10 organisms (Tchobanoglous et al. 2003), there is a significant risk of infection even after many log phase reductions in viral concentration.

Diarrheal diseases kill approximately 2.5 million people each year, the fifth highest cause of mortality worldwide (WHO 2008). Gastroenteritis, commonly caused by Rotavirus infections, accounts for 500,000 deaths and over 2 million infections per year in children under 5 years of age (WHO 2007). Over one-half of hospitalizations related to severe diarrhea in children and infants, are caused by Rotavirus infections.

Previous investigations of clay pot filters’ removal of viruses have been reported by van Halem (2006), Brown (2007) and Salsali et al. (2011) (Table 1). Large disparities in results between investigations are attributed to variances in methodology (source water characteristics and cleaning regime of the CWFs). There is also the potential for variance in methodology (source water characteristics and cleaning regime of the CWFs). There is also the potential for variance in methodology (source water characteristics and cleaning regime of the CWFs). There is also the potential for variance in methodology (source water characteristics and cleaning regime of the CWFs). There is also the potential for variation in materials and construction methodology.

Salsali et al. (2011) demonstrated the ability of CWFs to remove viruses from de-ionized water while employing a frequent (daily) cleaning regime. When compared to other studies that employed less regimented cleaning regimes (van Halem 2006; Brown 2007), removal efficiency of viruses was low. Frequent cleaning inhibits microbiological growth on the interior surface of the CWF and dislodges particulate matter, reducing CWF pore obstruction.

Pore obstruction, caused by the build-up of particulate matter, decreases the effective pore diameter of the filter element, resulting in increased viral removal efficiency due to more effective filtration. Microbiological/biofilm growth may also increase the attenuation of viruses. Hijnen et al. (2004) reported on the interaction of MS2 bacteriophages and biofilms (termed Schmutzdecke) during slow sand filtration. Phage reduction was determined to be between 1.5 and 2 log; however, this reduction was not due to the interaction of phage and biofilm alone, as mechanical filtration and adsorption processes occur throughout the sand bed.

Lantagne (2001) found that scrubbing the interior surface of the CWF with a brush removed many of the particles on the inner filter wall and re-established flow-through rates to within the manufacturer’s range. The results reported in Salsali et al. (2011) indicated low viral removal when compared to the investigations by van Halem (2006) and Brown (2007), both of whom utilized surface water as an influent source as well as a less regimented cleaning regime than adopted in Salsali et al. (2011).

Source water turbidity and cleaning regime were hypothesized (by all of van Halem 2006; Brown 2007; Salsali et al. 2011) as major contributors to the variation in data between experiments. This hypothesis is a plausible explanation for the discrepancies indicated in Table 1. Of interest herein is to extend the assessment reported in Salsali et al. (2011) and more fully identify the variables influencing virus removal during CWF filtration processes.

**Materials and Methods**

Two types of CWF were used for testing and were sourced from suppliers in Cambodia (Resource Development International Cambodia (RDIC) and International Development

<table>
<thead>
<tr>
<th>Researcher</th>
<th>Cleaning regime</th>
<th>Influent source</th>
<th>Turbidity (NTU)</th>
<th>Phage reduction (log phase units)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Salsali et al. (2011)</td>
<td>Strict, interior surface of pot scrubbed daily</td>
<td>De-ionized water</td>
<td>0.5–1.5</td>
<td>0.2–0.5</td>
</tr>
<tr>
<td>van Halem (2006)</td>
<td>Cleaning only in weeks 6 and 9 (13 week test interval)</td>
<td>Surface water (Canal Schie, The Netherlands)</td>
<td>Not stipulated</td>
<td>0.5–3</td>
</tr>
<tr>
<td>Brown (2007)</td>
<td>Cleaned once per week</td>
<td>Surface water (Cambodia)</td>
<td>8.4 (average)</td>
<td>1–2 (mean 1.3)</td>
</tr>
<tr>
<td>Brown (2007)</td>
<td>Cleaned once per week</td>
<td>Rain water (Cambodia)</td>
<td>1.1 (average)</td>
<td>1–2 (mean 1.4)</td>
</tr>
</tbody>
</table>
Enterprises (IDE)). Both CWF types received an application of colloidal silver solution as part of their manufacture, to act as a biocide. The primary difference between the two CWF types is that in the case of IDE, 4% laterite is mixed with the dry ingredients prior to firing (van Halem 2006). Laterite, a soil type rich in iron oxide, is hypothesized to aid in the removal of bacteria and heavy metals (Brown 2007). The CWFs were shipped to Guelph for testing; plastic receptacles (polyethylene rain barrels), obtained locally in Guelph, Ontario, were employed for collection of effluent. The effluent was thoroughly mixed before sampling. Between successive experiments, the plastic receptacles were cleaned with alcohol, and then rinsed with deionized water. All experiments were carried out at an ambient temperature of 22 °C.

Influent and effluent MS2 bacteriophage (ATCC: 15597-B1) concentrations were determined utilizing the double agar layer (DAL) method (USEPA 2001). Clear viral plaques in an opaque bacterial lawn were used to quantify coliphage concentration. Serial dilutions (10⁻¹ to 10⁻⁷) were plated in accordance with the DAL procedure to obtain a countable number of viral plaques (10⁴–400). Influent samples were collected after mixing to account for any turbulence-related reduction in phage concentration. The host bacterial stock (used for enumeration) was an antibiotic resistant Escherichia coli (E. coli Famp ATCC: 700891).

Cleaning methodology and influent parameters have been evidenced to have a large impact on phage removal efficiency of CWFs. The trials reported herein employ variations in source water turbidity and cleaning regime to identify the mechanisms responsible for phage removal during CWF filtration. To obtain source water with different levels of turbidity, de-ionized water was spiked with lab-grade bentonite clay. For all turbid trials, influent samples were taken before bentonite addition. Bentonite is an inexpensive, absorbent clay suitable for addition to CWFs in developing countries. Extrapolation of the results reported herein to removal efficiencies of CWFs in the field would be based on the components of source water turbidity. Turbidity derived solely from clay would result in a larger degree of viral sorption than turbidity that is composed of both organic and inorganic constituents.

CWFs employed in this study were previously investigated by Salsali et al. (2011). Using the same pots for testing allows detailed comparisons between results, due to the fact that pots from the same manufacturer will have some level of variability with regard to microbial removal efficiency. One CWF of each type was employed for testing to compare the effects of cleaning regime and source water turbidity independently of variations present between CWFs of the same manufacturer. Data distribution was tested using the Shapiro-Wilks test of normality. Log-transformed data were also tested for normality. Statistical significance testing two data sets of unequal variance was carried out using Satterthwaite’s modified t-test (McBean & Rovers 1998).

Effects of turbidity on CWF phage removal efficiency

To assess the effects of influent turbidity on CWF phage removal efficiency, de-ionized water was spiked with bentonite clay (1.0 ± 0.1 g/L) to obtain an influent turbidity within the range of 6–8 NTU. Trial duration was 3 h, and effluent volumes were monitored every half hour. Flow rates ranged from 1.5 to 2.5 L/h. Flow rates occasionally decreased slightly as the trial progressed due to accumulation of particles on the interior surface of the CWF and reduced hydraulic head within the CWF. Samples were taken after the three-hour test interval for plating in accordance with the DAL procedure. After trial completion, CWFs were thoroughly cleaned (interior surface washed and scrubbed) to reduce retained particles and improve repeatability between trials. The average influent coliphage concentration for all experiments was 1.2 × 10⁵ pfu/mL. In comparison to viral concentrations in the environment, high influent viral concentrations were used during experiments to improve statistical significance between experimental regimes. Influent and effluent samples were exposed to ambient temperature conditions for the same duration, to ensure that temperature-related viral inactivation did not have an impact on the data.

Effects of sorption on phages

Bentonite clay was mixed with a sample of MS2 (concentration 1.5 × 10⁵ pfu/mL) to obtain various turbidities (14.9 NTU, 7.5 NTU, 3.7 NTU). All samples were produced from the same virus-spiked stock, to ensure equal initial concentrations at all turbidity levels. The samples were then centrifuged (2800 rpm, 30 min) and the supernatant collected. Turbidity levels were determined both before and
after centrifuging to ensure removal of bentonite clay from the supernatant. This allowed the effect of sorption to be quantified. In all trials supernatant turbidity was <1 NTU. The initial sample and supernatant sample were plated in accordance with the DAL procedure (USEPA 2001).

**Effects of pore obstruction on CWF phage removal efficiency**

To investigate the effects of partial pore obstruction, 100L of highly turbid water (approx. 90 NTU) was filtered through each CWF over a period of 14 days. Average flow rate and turbidity data were recorded each day over the 14-day period. After clogging (as a result of clay addition), the CWFs were assessed for phage removal efficiency in accordance with the DAL procedure (USEPA 2001).

**RESULTS AND DISCUSSION**

**Effects of turbidity on CWF phage removal efficiency**

The variations in average log removal of the two CWF types are depicted in Figure 2. Data distribution was determined to be log-normal using the Shapiro-Wilks test. The literature indicates that the Shapiro-Wilks test performs well on small sample sizes, in comparison to other statistical tests of normality (McBean & Rovers 1998). During turbid influent trials, effluent turbidity was approximately 1 NTU on average, indicating both CWF types filter a large majority of clay from the influent. The increase in virus removal efficiency, as apparent in Figure 2, is the result of sorption of the phage onto clay particles, and subsequent filtration of the clay particles by the CWF. This finding was further demonstrated by investigation of viral sorption to clay particles at various turbidities.

Statistical testing (using Satterthwaite’s modified t-test) was undertaken to establish the degree of statistical significance between data sets. Data were compared on the basis of influent water turbidity (<1 NTU vs. 6–8 NTU), as well as CWF manufacturer (RDIC vs. IDE).

Increased levels of significance indicate increased likelihood of independence between sample groups. Comparative results indicate there is a statistically significant difference ($P < 0.05$) in phage removal efficiency between CWFs with turbid influent source water (6–8 NTU) and those with clear (<1 NTU) influent source water in the case of CWFs manufactured by RDIC. In contrast, phage removal efficiencies for CWFs manufactured by IDE were not significantly affected by clay addition within the range of turbidities used in this experiment.

Statistical comparison of CWF types (RDIC vs. IDE) during turbidity trials (6–8 NTU influent turbidity) indicated a low level ($P < 0.5$) of confidence regarding statistical independence. Comparison of CWF types (RDIC vs. IDE) during clear influent trials demonstrated a higher degree of confidence ($P < 0.1$), in comparison to turbidity trials. These results indicate that CWFs manufactured by IDE may be superior with respect to viral removal efficiency at low turbidity levels. This could be due to the presence of laterite in CWFs manufactured by IDE. Brown (2007) reported laterite to have a negligible impact on removal efficiency but did not investigate clear influent water sources. To accurately assess the impact of laterite on CWF viral removal efficiency larger data sets are required with multiple CWFs from each manufacturer. CWFs from the same manufacturer can have a range of characteristics due to variances in production practices.

**Effects of sorption on phages**

Figure 3 indicates a phage concentration reduction of 0.4–0.5 log when bentonite clay is added to the solution. Significant
viral sorption occurs even with low bentonite doses (<4 NTU). This finding indicates that CWF performance may be improved by utilizing influent source water with bentonite clay added, as compared to clear (<1 NTU) influent source water. It is noted, however, that variances between the investigations reported herein and other studies utilizing surface water as an influent may be due to the composition of source water turbidity, and the potential for aggregation of influent viruses. Suspended solids, phytoplankton and humic acids all influence turbidity. When bentonite clay is the only source of turbidity, a large degree of sorption occurs. The degree of sorption evidenced herein may vary from the degree of sorption achieved by other components of turbidity (e.g. humic acids, phytoplankton), which may affect the efficiency of viral removal. In addition, viruses in the environment are commonly shed with faeces and may occur naturally sorbed to other particles. This phenomenon may decrease the effectiveness of bentonite addition in practice. In instances when viruses are naturally aggregated, they are more easily removed during filtration (in comparison to free viruses), and will be less likely to sorb to bentonite particles. Therefore, the effectiveness of bentonite addition will be reduced.

Effects of cake layer formation on CWF phage removal efficiency

Although sorption was observed to have an effect on CWF removal efficiency, the process of sorption alone did not account for the discrepancies between Salsali et al. (2011) and Brown (2007) due to variations in cleaning regime and test duration. Lantagne (2001) found that clogging significantly reduced flow rates if the CWFs were not cleaned regularly.

During cake layer formation trials, average influent and effluent turbidities were 93 and 1 NTU respectively. As with previous tests, the majority of clay entering the CWF is retained on the filter element. It is interesting to note, however, that allowing the CWF to dry over a period greater than two days, noticeably decreases the flow rate of subsequent tests as apparent between the dates of 28 July and 3 August in Figure 4. The decreased flow rate is hypothesized to be the result of hardening of clay particles within CWF pore spaces. As the moisture between bentonite particles evaporates, the structure solidifies, blocking the largest pores of the CWF and resulting in a smaller average pore size. This process is expected to be irreversible and subsequent addition of moisture does not cause the clay to redisperse in solution over the short term (1–2 weeks). Long-term exposure of the clay cake layer to moisture may cause
re-dispersion in solution; however, this process requires further investigation.

After establishing a clay cake layer as a result of the highly turbid influent source water, the CWFs were again tested for viral removal efficiency. Five trials were investigated, following the same procedure as outlined in the Materials and Methods section. The results depicted in Figure 5 demonstrate that the formation of a clay cake layer within the CWF drastically increases phage removal efficiency. This phenomenon is hypothesized to be the result of increased sorption onto clay particles within the CWF pore spaces. These findings indicate that decreasing the frequency of cleaning improves CWF viral removal efficiency. However, decreasing the frequency of cleaning also decreases flow rate during CWF operation. The trials reported herein evidence a reduction in flow of approximately 50%, in comparison to clear influent trials. While this reduction in flow rate is acceptable (still within manufacturer’s specified range), continued usage over the long term may cause further decreases in flow rate and necessitate cleaning. The authors recommend a less intensive cleaning regime than currently adopted by CWF users.

If a viral outbreak was identified within a population, bentonite clay could be added to potable water prior to filtering, as a cost-effective method of improving viral removal efficiency of CWFs. However, if the viral outbreak caused many log increases in influent viral concentration, bentonite addition may have too small an effect on effluent quality to reduce infection rates.

The presence of bacteria in influent source water may also impact viral removal efficiency of CWFs. If a substantial fraction of enteric viruses sorb to the surface of bacteria, viral removal efficiency will be improved. Bacteria are efficiently removed from source water as a result of filtration processes (size exclusion), similar to the removal of clay particles. Murphy et al. (2010) reported bacterial removal efficiencies of CWFs from 2 to 4 log depending on influent bacterial concentrations.

**CONCLUSIONS**

Investigations of virus removal efficiency of two (IDE, RDIC) CWFs under various experimental regimes indicated slight increases in virus removal efficiency with the addition of bentonite clay, and substantial increases in viral removal when using CWFs with an established clay cake layer. Utilizing an influent water source with bentonite clay evidenced...
increases in viral removal efficiency of CWFs by 0.1 log (IDE) and 0.2 log (RDIC), when compared to de-ionized influent source water.

Using a filter with an applied clay cake layer provides improved phage removal efficiency over both clear influent and turbid influent trials. However, formation of a clay cake layer also had an adverse effect on flow rate, causing a 50% flow reduction, in comparison to a clean CWF. Log reductions of 1.9–2.5 were obtained during trials which employed clay cake layers; compared to values of 0.2–0.5 for trials involving a de-ionized influent and frequent CWF cleaning.

RECOMMENDATIONS FOR FUTURE RESEARCH

CWF viral removal efficiency may be improved during normal use by the following alterations, all of which require additional investigation and evaluation:

1. Decrease the frequency of cleaning regime thus allowing the CWF to naturally establish a cake layer or biofilm.
2. Pre-coat the interior surface of the CWF with bentonite clay during manufacture. This alteration may have an adverse effect on flow rate.
3. Investigate phage removal in combination with the addition of influent bacteria. Viruses may sorb to bacteria similarly to clay particles and thus be more efficiently removed.

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