Evaluation of a silver-impregnated ceramic filter for small-scale rainwater purification using viral and bacterial indicators

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Abstract: The purpose of this work is to examine the potential of a cost-effective technique for purifying rainwater collected for human consumption. The technology consists of a ceramic cartridge impregnated with colloidal silver developed in Mexico, which can be attached to a conventional drinking water dispenser. Although abundant information exists on the uses of silver for water purification, little is known about its application for small-scale RWH systems, where specific concerns prevail, such as long-term storage (i.e. throughout the dry season) which increases the risk of stagnation and contamination. The purifiers are currently being evaluated in laboratory experiments and field tests to better understand the mechanisms involved. Among the parameters being used to test disinfection efficiency, a technique for determining levels of viruses (phages) capable of infecting E. coli has been included, which may provide a better assessment of risk (particularly from viral pathogens) when compared with traditional bacterial indicators. Variation in chemical composition of the water throughout the filtering process is also analysed. Once the scope and efficiency of this technology is determined it is envisaged that it may become a valuable tool for ensuring adequate rainwater quality at an affordable cost for households and communities in developing countries.

Keywords: Colloidal silver, Rainwater harvesting, Ceramic filtration, Bacteriophages, viral indicators

1. Introduction

Access to clean drinking water, with the associated impacts on health and overall quality of life, is a matter of increasing worldwide concern, particularly for developing countries. It is estimated that close to 2,000 children die every day from preventable water-related diseases [Gleick 2014; Water Aid 2014]. Sustainable water sources and feasible treatment methods are essential in order to resolve this crisis. Rainwater Harvesting (RWH) can be considered in many cases as an attractive alternative supply due to its relatively low concentration of hazardous contaminants such as arsenic, manganese, and fluoride, as well as low hardness [Banks & Heinichen 2006; Malik et al. 2003]. However, rainwater collected from rooftops or other surfaces rarely attains potable water standards unless some sort of treatment and/or disinfection is applied [Gould. et al. 1999]. In this context, Ceramic Water Filters (CWF) coated or impregnated with silver can be used as a low cost and effective point-of-use water purification technique, providing both filtration and disinfection functions.

Some of the important advantages of CWFs are their relatively high porosity, ease of installation and maintenance, coupled with the fact that they are gravity-driven and thus do not require external energy to operate [Simonis & Basson 2011]. Moreover, low costs (around USD $0.2-$0.3 each) and relatively mature manufacturing technologies have made CWFs widely adopted around the world [Hasan et al. 2011; Laan et al. 2014]. Its purification function is mainly achieved by filtration, due to small pore sizes, combined with electrostatic adsorptive forces which also facilitate removing certain particle sizes. In some cases, CWFs are impregnated with silver to further enhance microorganism removal effects.

Previous studies show the efficiency of CWFs in removing physical particles and bacteria. Bielefeldt [2010] found removal efficiencies to be generally above 99.6% when testing different particle sizes (1 - 10 μm). It was found that this efficiency increased slightly as the experiment continued, which could be attributed to the formation of a biofilm layer on CWFs' surfaces. Applying silver would have little impacts on overall turbidity removal efficiencies, and it may even pose negative effects on turbidity removal in the longer term since silver would presumably prevent biofilm formation [Oyanedel-Craver & Smith 2008; Bielefeldt et al. 2009; van Halem et al. 2009]. Other studies have found CWFs to have highly unstable outputs, sometimes as low as 88% [Simonis & Basson 2011; Hwang 2003; Sobsey 2002]. Applying silver has been shown to mitigate this limitation by driving bacterial removal efficiencies consistently above 98% [Dies 2001; McAllister 2005; Baumgartner 2006; Simonis & Basson 2011]. However, CWFs performance in the elimination of viral surrogates (i.e. bacteriophages) have thus far been shown to be relatively poor and inconsistent, with highly variable results. Simonis & Basson [2011] found a Log Reduction Value of Bacteriophage (LRVP) between 0.21 and 0.45 using Deionised Water (DW) as media, while Oyanedel-Craver & Smith [2008] also got low virus removal efficiencies, well below 90%. Other studies using microspheres [of size 0.02um - 0.1μm] as viral surrogates, found a highly variable LRVP from 0.43 to 2.4 [63% - 99.3%] when tested with six filters [Bielefeldt et al. 2010]. Moreover, it was also seen that silver posed little improvement in phage elimination [Van der Laan 2014].

The abovementioned experiments had limitations, as a media of relatively clean water was mostly used and silver was generally applied by painting or coating external ceramic surfaces. In the present study, a different type of CWF (where colloidal silver is directly impregnated during the manufacturing process) has been tested using rainwater as a media. The key object of this research is to experimentally analyze and compare the phage elimination capabilities achieved by both n-CWF (ceramic filters without colloidal silver) and cs-CWF (with colloidal silver). The disinfection effect of colloidal silver was assessed by setting contrast experiments. Meanwhile, filtration rates and the ability to remove inherent physicochemical particles as well as bacteria were also assessed.

2. Methodology

Four new CWFs were obtained from a Mexican NGO with ample experience in their manufacture and implementation in the field [CATIS Mexico 1994], two impregnated with colloidal silver (200 ppm each) and the other with the purely ceramic composition, without the silver. These were assembled at the Environmental Engineering lab of University College London (UCL) using identical setups, as shown in Figure 1. A basic description of the four CWF assemblies can be seen on Table 1.

Table 1: CWF assembly details

Assembly Code	Condition		Purposes
S1	With Silver	Brand New	Bacterial Removal Experiment
S2	With Silver	Brand New	Virus Removal Experiment
N1	No Silver	Brand New	Blank of 'S1'
N2	No Silver	Brand New	Blank of 'S2'

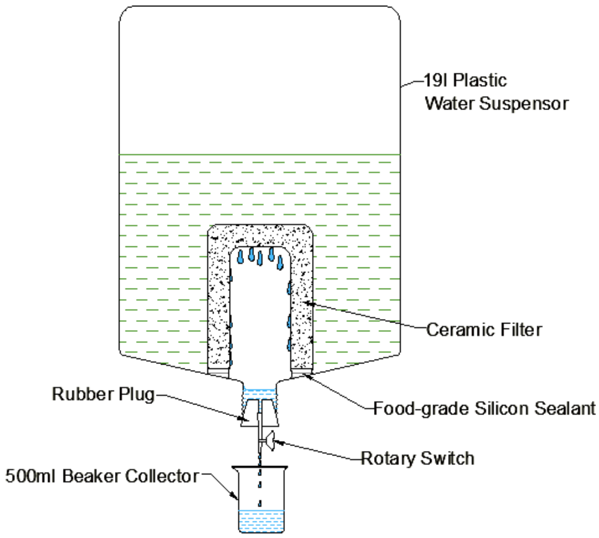


Figure 1: CWF experimental setup

2.1 General parameters

Filtration rates were achieved from the quotients of 'volume to time', which were recorded as periods of time spent in collecting each 100ml aliquot samples. Meanwhile, water column heights in feeder tanks (suspensors) were also recorded simultaneously. Regarding water quality, the main physico-chemical parameters that were tested and correlated are highlighted in Table 2.

Table 2: Testing specifications of general physico-chemical parameters

Parameter	Test Method	Parameter	Test Method
pH	Jenway 4330 Conductivity & PH meter	Turbidity	Hach Turbidimeter
Conductivity		Silver ions	Hach DR2800 Spectrophotometer
Total Dissolved Solid [TDS]		TOC [Total Organic Carbon]	Shimadzu TOC-L Analyser
Temperature		Cations/Anions	Dionex ICS-1100 Ion Chromatograph

2.2 Bacterial Removal Tests

E. coli (strain ATCC 11775) was used to spike water for testing bacterial removal efficiencies in the filters. These were prepared by incubation on Tryptic Soy Agar (TSA) for 24h at 36 ± 2°C, after which the colonies were scraped off and diluted into 10% PBS (Phosphate Buffer Solution). For each experiment, 10ml of this concentrated solution ($\approx 10^8$ CFU/ml) were added into each feeder tank, containing approximately 8L of rainwater, followed by gentle stirring to ensure adequate mixing. Samples (100ml) were collected both from influents and effluents when water column heights (WCH) in the feeder tanks had dropped to 7.5L and 5L respectively (to demonstrate how efficacy would vary against different flow rates). These were analysed using dilutions as required and incubated in HACH m-ColiBlue24® broth, according to the membrane filtration method [Crane et al. 2006].

2.3 Phage Removal Tests

To model the potential for virus removal in the filters, somatic coliphages were used as surrogates, according to an emerging technique (EN ISO 10705-2:2001). These are considered safe and relatively easy to enumerate. Host bacterial strains (WG-5 E. coli) were incubated into 25ml of Modified Scholtens Broth (MSB) for 24h at 36 ± 2°C while shaking at 110 rpm, and prepared according to the standard method cited above. The initial stock of host bacteria and coliphages were obtained from the Environment & Technology Dept. at the University of Brighton. Throughout the duration of the experiments, phage solutions were stored in 2ml vials at(5 ± 2)°C. Phage density in these solutions was detected to be around 1.3×10^9 pfu/ml. The WG-5 host bacteria was stored separately in a -80°C freezer, also in 2ml vials.

WG-5 Phage Spike

Inoculate a vial of WG-5 phage stocks (1.3×10^9 pfu/ml) into suspensor containing 10L rainwater.

Sampling

Samples (100ml] would be collected both from influents and effluents when water column heights (WCH] in suspensors were at 7.5L and 5.5L respectively.

Sample Preparation

Pass raw samples through 0.22μm filters and collect filtrates. Then dilute samples to 10-3 or 10-4 for influent and 10-2 or 10-3 for effluent.

Inoculum Cultures Preparation (WG-5 E.coli]

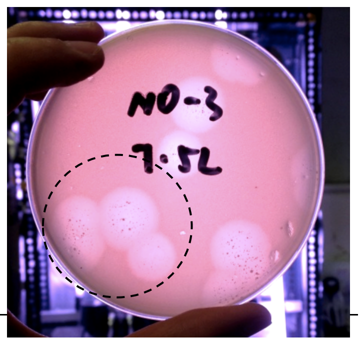
Remove a vial of prepared WG-5 E. coli stock from freezer and equilibrate to room temperature.
Add into 50ml MSB and incubate at (36 ± 2)°C with 110rpm shaking.
Grab 3ml samples every 30mins or 1 hour and analyse using spectrophotometer. Cease incubation until it reaches 0.333 Abs *(Corresponded to*10^8 *cfu/ml]* and then place in melting ice. *(Incubation generally takes 2.5h to 3.5h)*

Plate Assembling

Mix 1ml sample, 1ml inoculum culture, 300μl calcium chloride, and 2.5ml ssMSA together, and then evenly pour mixtures on MSA plates.

Incubation & Plague Counting

Incubate MSA plates upside-down at $(36 \pm 2]$°C for(18 ± 2)h. Count numbers of plaques appeared on each plates.



2.4 Minimum Inhibitory Concentration

To test the silver concentrations at which phages would be inactivated, an independent experiment was performed using a Hach silver standard solution (1000∓10mg/l). 1ml of WG-5 Coliphage stock was spiked into a feeder tank (suspensor) containing rainwater. Subsequently, four vials (60ml each) with different silver concentrations were prepared using ultra-pure Milli-Q water to dilute the silver standard solution accordingly (achieving concentrations of 50, 100, 150 and 200 respectively). A blank was also prepared. Furthermore, 3ml of phage-containing sample were added to these silver solutions, with a 15 minute reaction time. Finally, samples were passed through 0.22µm pore-size filters, using 1ml from each filtered sample to perform the phage analysis according to the method described in the previous section.

3. Results & Discussion

3.1 Physico-chemical parameters

Flow rates
Filtration rates were observed to vary between 1L/h and 3L/h, with a strict correlation observed with column heights, which varied from about 15 to 25 cm for the various experiments performed. As the water column continued to drop within any specific test, flow rates were reduced to virtually zero.

pH
Marked increases in pH values could be observed after both deionised water and rainwater were passed through the CWFs. The strong alkalinity of effluents during the first runs (Figure 2) exceeded the 8.5 pH upper limit of potable water standards [WHO 2011]. According to the manufacturer [CATIS Mexico 1994], one of the potential factors leading to high pH could be the water used during production and rinsing processes, which was highly alkaline, as well as the source materials used (i.e. clay). pH values tended to stabilise after subsequent flushes.

Turbidity
Rainwater was relatively clean, with turbidity ranging from 0.6–1.6 NTU. The CWFs showed excellent turbidity removal efficiencies with effluents stabilising between 0.1–0.2 NTU after a first rinse of 20L. CATIS also conducted similar tests using high turbidity influents (>400NTU), showing similarly results, with the effluent within 0.71 – 0.92 NTU [CATIS Mexico 1994].

Conductivity
High conductivity was observed in the effluents (Figure 3). This could be attributed to impurities and residues originally present on the CWFs, as well as clay particles. Though conductivity tended to stabilise after subsequent flushes, effluent values were consistently higher than influent values, possibly due to the dissolution of calcium and other chemicals upon contact with the ceramic surface.

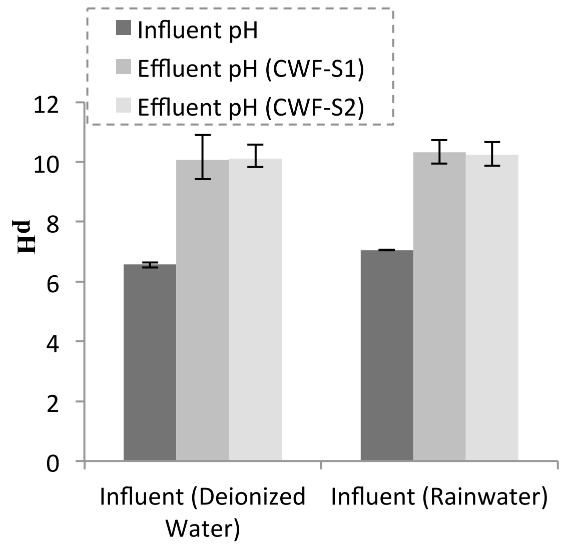


Figure 2: Average pH Values

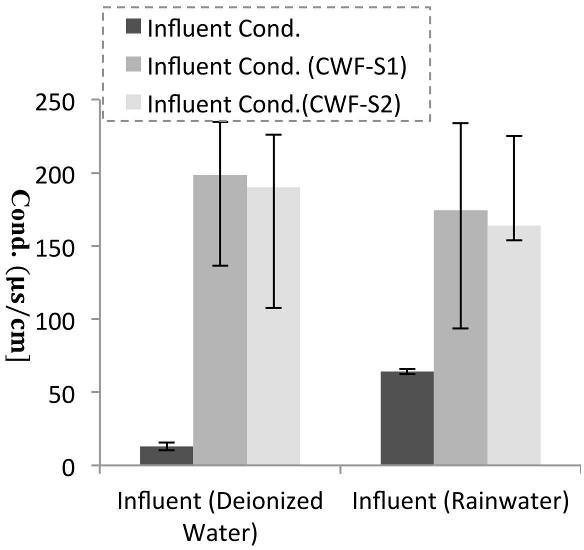


Figure 3: Average Conductivity Values

3.2 Phage Removal Test

Results from phage removal tests are shown in Table 3. Phage removal efficacies (LRV_P) of n-CWFs (without colloidal silver) were found to be between 0.21 – 0.67, while the LRV_P values achieved by cs-CWFs (impregnated with colloidal silver) were higher and fell between 0.52 – 1.24. Statistical t-tests were performed in order to assess these efficiencies, with results shown in Table 4.

Table 3: Phage removal test results

Batch	Sample Point when WCHs at [L]	Normal CWF				Colloidal Silver CWF			
		Influent Phage Conc. [pfu/ml]	Effluent Phage Conc. [pfu/ml]	% Red.	LRV	Influent Phage Conc. [pfu/ml]	Effluent Phage Conc. [pfu/ml]	% Red.	LRV
8	7	3.20×10^4	8.00×10^3	75.0%	0.60	1.10×10^4	2.80×10^3	74.5%	0.59
9	7.5	4.90×10^4	1.15×10^4	76.5%	0.63	3.14×10^4	3.13×10^3	90.0%	1.00
9	4.5	4.00×10^4	1.05×10^4	73.8%	0.58	2.10×10^4	1.20×10^3	94.3%	1.24
10	7.5	5.85×10^4	1.30×10^4	77.8%	0.65	3.90×10^4	6.00×10^3	84.6%	0.81
10	5.5	3.20×10^4	1.30×10^4	59.4%	0.39	1.90×10^4	2.80×10^3	85.3%	0.83
11	7.5	7.55×10^4	1.60×10^4	78.8%	0.67	6.15×10^4	1.50×10^4	75.6%	0.61
11	6.5	2.95×10^4	1.80×10^4	39.0%	0.21	1.65×104	4.95×10^3	70.0%	0.52
12	7.5	1.20×10^5	3.30×10^4	72.5%	0.56	1.00×10^5	2.35×10^4	76.5%	0.63

Table 3: Summary of t-tests of phage test results

Comparisons	t stat	t-critical	P [t-test]
Phage concentrations in influents & effluents [n-CWF]	3.493	2.306 [Two-tail]	P [two-tail] =0.00816
Phage concentrations in influents & effluents [cs-CWF]	2.742	2.306 [Two-tail]	P [two-tail] =0.0253
Phage concentrations in influents from n-CWF & cs-CWF	1.129	1.761 [One-tail]	P [one-tail] =0.1389
Phage concentrations in effluents from n-CWF & cs-CWF	2.046	1.761 [One-tail]	P [one-tail] =0.03
LRVs of n-CWF and cs-CWF	2.363	1.782 [One-tail]	P [one-tail] =0.0179

Despite significant reductions in phage concentrations, there was still a difference between the achieved LRV_P values and WHO guidelines for microbiological disinfection (LRV>3). Unlike the case of bacteria, the pore sizes of CWFs are too large (0.6 – 3 μm) to retain viral particles (<0.2 μm), even though some retention does occur, as shown in the results. However, it is the addition of silver that accounts for the marked improvement in disinfection efficiencies.

LRVP and infiltration rates were in negative linear relationships (R2 = 0.99) as shown in Figure 3. An explanation for this finding is that when filtration rates were high, phages which may attach to the porous structures would be more easily sucked into high-velocity transient flows and passed into effluents. In the case of cs-CWF (with colloidal silver), filtration rates will directly affect the phage contact time with the silver particles, as can be observed by the steeper line of cs-CWF compared to n-CWF in Figure 4.

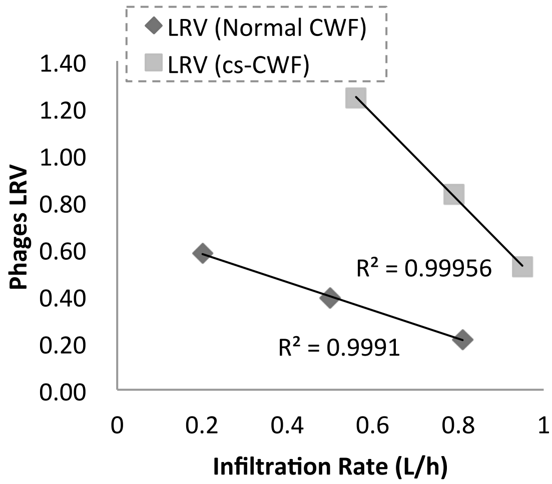


Figure 4: Phage removal efficiencies at different infiltration rates

3.3 Bacteria Removal Test (*E. coli*)

Results from bacteria removal tests are demonstrated in Table 4, which were comparable to the information stated by CATIS Mexico in their technical report, where it said their CWFs could eliminate 99.9% (LRV=3) of bacteria [CATIS Mexico 1994]. This was confirmed by another independent laboratory which found a 99.73% (LRV=2.7) elimination rate of bacteria, when initial bacterial concentrations were 1100 CFU/100ml (ibid.).

Table 4: Bacterial removal test results

	WCHs in Suspensor	Influent E.coli [cfu/100ml]	Effluent E.coli [cfu/100ml]	% reductio n	Log Reduction Value [LRV]
n-CWF	6L	3.36×10^6	4.55×10^3	99.86%	2.87
	3L	3.36×10^6	2.76×10^3	99.92%	3.09
cs-CWF	6L	3.11×10^6	9	99.99%	5.56
	3L	3.11×10^6	2	99.99%	6.19

In addition, CWFs impregnated with colloidal silver (cs-CWF) attained better outcomes for bacterial removal (almost twice as those achieved by n-CWF). These results could be attributed to the silver ions released by the colloidal silver particles from cs-CWF. Finally, higher elimination efficiencies were found at lower water column heights (i.e. lower filtration rates), possibly due to increased contact and retention times as discussed previously. A similar phenomenon occurred during the phage tests (Figure 4).

3.4 Silver Test

The Minimum Inhibitory Concentration (MIC_{Ag}) in this context was tested as the ability of silver to completely eliminate phages after a contact time of 20 minutes. This time period was chosen so as to correlate results to a previous study done by Adler et al. [2012], which found the MIC_{Ag} to be 140ppb. For these experiments, silver dosing was prepared as explained in the methodology. Testing with a range of concentrations revealed that a total disinfection seems to occur between 90 and 130ppb Ag (Table 5). Comparing these results with the silver ion concentrations released into effluents by the CWFs, which were initially detected to be 0.04mg/L (400ppb) and gradually reduced to 0.01mg/L after filtering 100L rainwater, the presence of active silver ions appears to be far from sufficient to meet virus disinfection purposes (at the high phage concentrations being used here).

Table 5: Phage removal with different silver concentrations in effluents (after 20 min.)

Dilution \ Concentration of silver [ppb]	0 [blank]	40	90	130	180
0	TNTC	TNTC	----	0	0
10^{-1}	TNTC	22	9	0	0
10^{-2}	27	2	1	0	0
10^{-3}	6	0	0	0	-----
Average Conc. pfu/ml	8700	210	95	0	0
LRV_P compared to blank	0	1.62	1.96	n/a	n/a

In addition, it was also detected that silver concentrations actually achieved in rainwater were always below expected values (Figure 5).

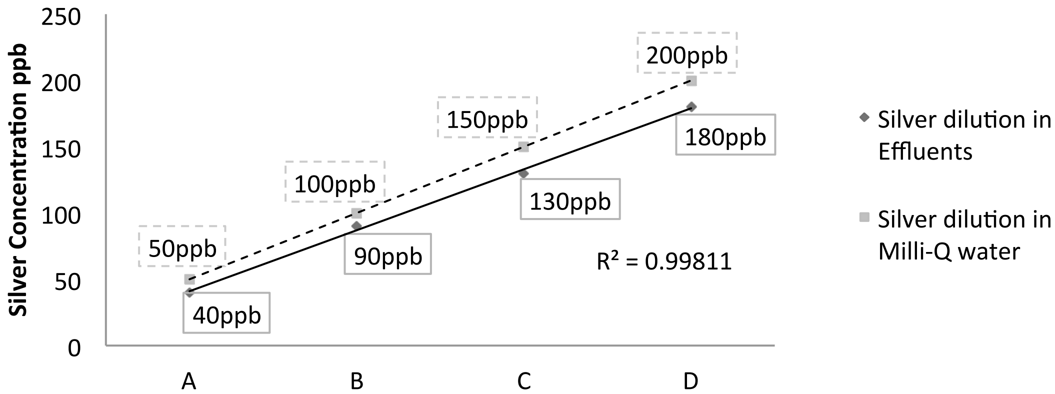


Figure 5: Silver concentrations from a diluted standard in Milli-Q & rainwater effluents

This could be due to the high pH of effluents, as well as interfering ions in the rainwater or in the filtering media which might complex and eventually precipitate silver ions [Adler et al. 2013], usually in the form of silver chloride, silver nitrate or other salts. Benjamin [2002] highlighted the insolubility of silver chloride, even though this could be mitigated by exposing silver chloride to light (Landau et al. 2007].

4 Conclusions & Recommendations

CWFs can generally be regarded as efficient water filters with the ability to supply a variable flow rate (up to 3 L/hour, in this case, linearly related to water column height). Without proper rinsing, however, impurities can accumulate in porous structures and lead to the formation of biofilms which can restrict flows. Turbidity removal efficiencies were shown to be consistently high, with effluents between 0.1 NTU and 0.2 NTU, well within any drinking water guidelines. Though conductivities were slightly increased after passing through CWFs, they were still within potable ranges.

The high alkalinity of effluents was shown to be a matter of concern, with the pH initially obtained above drinking water standards. This could be reduced after extensive rinsing (possibly over 100L of clean water), though the source of this alkalinity should be investigated further and analysed. The Log Reduction Values (LRV) of both *E. coli* and phages were in negative linear relationships with the filtration rates, meaning that the efficiency of the filter is enhanced when the water column is at its lowest (allowing ampler contact time with the silver). This has implications when considering more contaminated sources, where limiting column height might be used as a precautionary measure.

With regards to overall disinfection efficiencies, the main aim of the study, while satisfactory bacterial eliminations were achieved, the performance regarding viral indicators was not ideal. In both cases, the role of impregnated colloidal silver was confirmed to be crucial in enhancing these removal efficiencies, a fact which has been quite well known in the literature, though it was not present in high enough concentrations to effectively inactivate bacteriophages. Should virus be of concern to a particular water supply using CWFs for purification, it is likely that the silver dosing needs to be increased or further treatment applied.

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