

Journal of Water, Sanitation and Hygiene for Development
Development of improved low-cost ceramic water filters for viral removal in the Haitian context
 --Manuscript Draft--

Manuscript Number:	WASHDev-D-14-00121R1
Full Title:	Development of improved low-cost ceramic water filters for viral removal in the Haitian context
Short Title:	Improved low-cost ceramic water filters for viral removal
Article Type:	Research Paper
Corresponding Author:	Rosina Girones Laboratory of Virus Contaminants of Water and Food, Department of Microbiology, Faculty of Biology, University of Barcelona SPAIN
Corresponding Author's Institution:	Laboratory of Virus Contaminants of Water and Food, Department of Microbiology, Faculty of Biology, University of Barcelona
First Author:	Laura Guerrero-Latorre
Order of Authors:	Laura Guerrero-Latorre Marta Rusiñol Ayalkibet Hundesa Maite Garcia-Valles Salvador Martinez Osnick Joseph Silvia Bofill-Mas Rosina Girones
Abstract:	Household-based water treatment (HWT) is increasingly being promoted to improve water quality and, therefore, health status in low-income countries. Ceramic Water Filters (CWF) are used in many regions as sustainable HWT and have been proven to meet WHO microbiological performance targets for bacterial removal (2-4 log); however, the described viral removal efficiencies insufficient to significantly reduce associated risk to viral infection. With the objective of improving the viral removal efficiencies of ceramic water filters, new prototypes with different oxide compositions and firing atmospheres have been developed and evaluated. For removal efficiencies human adenoviruses, MS2 bacteriophage and E. coli were quantified in all prototypes. A new model of CWF that was fired in a reductive atmosphere presented virus and bacteria removal efficiencies greater than 3.0 log and 2.5 log, respectively, which would fulfill the viral targets that are recommended by the WHO. Ceramic characterization of the selected filters, which were fired in a reductive atmosphere, showed that a larger specific surface area than those of control filters and higher fraction of a positive Z-potential fraction are the most likely explanations for this increase in virus removal.
Additional Information:	
Question	Response
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Total word counts: **6176 words** (including tables and figures)

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4 2 **Development of improved low-cost ceramic water filters for viral removal in the**
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7 3 **Haitian context**
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14 5 Guerrero-Latorre L.¹, Rusiñol M.¹, Hundesa A.¹, Garcia-Valles M.², Martinez S.², Joseph O.³,
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16 6 Bofill-Mas S.¹, Girones R.¹.
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22 8 1. Laboratory of Virus Contaminants of Water and Food, Department of Microbiology, Faculty of Biology, University of Barcelona
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24 9 2. Department of Crystallography, Mineralogy and Mineral Deposits, University of Barcelona
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26

27 10 3. Laboratoire de Qualité de l'Eau et de l'Environnement, Université Quisqueya, BP 796 Port-au-Prince, Haiti.
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32 12 **Keywords:** Household Water Treatment and Safe Storage, Ceramic Water Filters, Waterborne viruses,
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34 13 Ceramic fired in Reductive Atmosphere. Viral Removal Efficiencies.
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41 15 **Abstract**
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46 17 therefore, health status in low-income countries. Ceramic Water Filters (CWF) are used in many regions as
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1	29	Abbreviations
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3	30	BET Brunauer, Emmett, Teller
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5	31	CFU colony forming unit
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7	32	CWF ceramic water filter
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10	33	<i>E.coli</i> <i>Escherichia coli</i>
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12	34	GC genomic copies
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14	35	HAdV Human adenoviruses
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16	36	HWT household water treatment
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19	37	ICAITI Instituto Centroamericano de Investigación y Tecnología Industria
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21	38	IEP Isoelectric Point
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23	39	IU International Units
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25	40	JCPyV JC Polyomavirus
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28	41	LRV log reduction value
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30	42	NoV (GI and GII) Norovirus (Genotype I and Genotype II)
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32	43	nPCR nested PCR
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34	44	PCR polymerase chain reaction
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37	45	PFU plaque forming units
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39	46	qPCR quantitative PCR
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41	47	RT-PCR reverse transcription PCR
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43	48	SD standard deviation
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46	49	WaSH water, sanitation and hygiene
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48	50	WHO World Health Organization
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50	51	XRF X-ray Fluorescence
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1 **52 Introduction**

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4 53 The World Health Organization estimates that 780 million people worldwide (42% in Sub-Saharan Africa)
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6 54 were still without access to improved sources of drinking water in 2010 (Onda, LoBuglio, and Bartram
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8 55 2012). Estimates of global young-child (<age 5 years) mortality from 2010 suggest that 2 million young
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10 56 children die due to diarrheal disease, and approximately 1.4 million (70%) of these are localized in low-
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12 57 income countries (Bryce et al. 2005; Black et al. 2010; Liu et al. 2012). Recent data on diarrhea burden in
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14 58 low-income countries has reported 685,000 diarrhea deaths related to the deaths attributable to inadequate
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16 59 water and sanitation in 2012 (Prüss-Ustün et al. 2014). The etiological origins of diarrhea had been
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18 60 traditionally associated with bacteria and protozoa; however, in the last years, improved diagnostic methods
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20 61 have made possible detection of other etiological agents, such as enteric viruses, which altered the
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22 62 distribution of the most common cause of diarrheal illness worldwide into enteric viruses (e.g., norovirus,
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24 63 rotavirus, adenovirus) (Huilan et al. 1991; Parashar et al. 2003; Kotloff et al. 2013).

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26 64 Large numbers of viruses are excreted in human feces and urine into the environment, and, even at low
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28 65 concentrations, they can cause illness when ingested (Leclerc, Schwartzbrod, and Dei-Cas 2002).
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30 66 Consequently, sewage and fecal contamination, if not well contained and treated, is the main source of
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32 67 microbiological risk in water and food. In countries with poor sanitation systems, such as in Haiti, where
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34 68 only 17% of the population has improved sanitation (Onda, LoBuglio, and Bartram 2012), microbiological
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36 69 contamination in water sources is a major cause of morbidity and mortality.

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38 70 Most potable water-treatment methods are not suitable or available for the majority of rural areas in low-
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40 71 income countries without reliable access to safe drinking water. For those populations, household water
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42 72 treatments (HWT) have been promoted to improve water quality and prevent diarrhea because of two
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44 73 important characteristics: low cost and the capacity, from some of them, to be produced locally. Among all
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46 74 of the technologies tested at the laboratory and field levels, ceramic water filters (CWF) have shown the
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48 75 highest impact on long-term health due to the high adherence of users and their capacity to reduce
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50 76 approximately 50% of the diarrhea incidences (T. F. Clasen et al. 2004; T. Clasen et al. 2005; Joe Brown,
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52 77 Sobsey, and Loomis 2008; Preez et al. 2008; Hunter 2009; Levine, Ave, and Francisco 2010).
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1 78 Currently, the most widely available, locally produced model of CWF is based on a design developed in
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3 79 1981 by the Guatemalan industrial research institute ICAITI (*Instituto Centro Americano de Investigación*
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5 80 *y Tecnología Industrial*) and recently updated by The Ceramics Manufacturing Working Group (The
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7 81 Ceramics Manufacturing Working Group 2011)

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10 82 The performance of this CWF with and without colloidal silver has been investigated over the years by
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12 83 researchers who have studied its efficiency at reducing waterborne pathogens in the laboratory and in the
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14 84 field. In the literature, the removal efficiency of bacteria by CWF is described to be within the range of 2 to
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16 85 4 logs using the standard types of silver-impregnated CWFs (Halem 2006; Oyanedel-Craver and Smith
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18 86 2008; The Ceramics Manufacturing Working Group 2011); however, viral removal efficiencies have been
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21 87 shown to be much lower, between 0.21 and 0.45 log in CWF impregnated or not impregnated with silver
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23 88 described by Salsali H, McBean E 2011 study, and around 1.5 log reduction in Brown & Sobsey 2010 study
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25 89 with no significant differences between impregnated and non-impregnated filters.

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28 90 On the other hand, the levels of viral removal by the actual model of CWF would not be effective in
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30 91 decreasing the microbial risk regarding the prevalence and concentration of viruses in surface waters, which
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32 92 has been recently reviewed and showed a prevalence as high as 2-4 logs/L of human adenoviruses (HAdV)
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34 93 in surface waters from different geographical areas worldwide (Bofill-Mas et al. 2013).

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37 94 Regarding the question of viral retention/inactivation in clay structures, different approaches have been
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39 95 presented in the literature, which have faced the challenge of viral removal from water:

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41 96 A substantial body of literature on virus attachment to various, positively charged particles and surfaces,
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43 97 especially metal oxides and positively charged media in the environment such as clays, which are rich in
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45 98 ferric oxides, exist (Landry et al. 1979; Gerba 1984; Loveland et al. 1996; Zhuang and Jin 2003).

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48 99 Brown & Sobsey (J Brown and Sobsey 2009) found that metal oxides such as α -FeO(OH) (goethite), Fe₃O₄
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50 100 (magnetite), Fe₂O₃ (hematite) and Al₂O₃ (alumina) that were amended to clay material could effectively
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52 101 capture bacteriophages (up to 8 log) from a solution, although no assays were performed using real filters.
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54 102 Moreover, ceramic depth filters with added Magnesium Oxide (MgO) showed an improved viral removal,
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57 103 however, the experiments presented high variability in performance with filter operation time, being
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59 104 discarded for drinking water production (Benjamin Michen et al. 2013).

1 105 Other factors related to hydrophobic interactions, surface environment and chemistry could also be
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3 106 important in virus-media adsorption and inactivation. It also has been described in the context that firing
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5 107 ceramics in reductive atmosphere has an effect on the surface charge of the piece and therefore can have an
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7 108 effect on viral retention (Wegmann, Michen, and Graule 2008). The shift of the IEP into positive charge at
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9 109 pH 7 makes this material interesting for being used in a virus adsorption filter considering the fact that most
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11 110 enteric viruses have a net negative surface charge (B Michen and Graule 2010).

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14 111 We hypothesized that the efficiency of the removal of viruses from water using CWF might be increased by
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16 112 using CWFs that were produced with clays amended to natural oxides (Fe₂O₃, Fe₃O₄, Al₂O₃), which
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18 113 increase the positive charges. Moreover, clays from Girona (Spain) and Haitian local clays were used to
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20 114 create CWF by firing in a kiln with a reductive atmosphere, which might affect viral retention as described
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22 115 previously.

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25 116 This study has as a main objective to design a CWF applicable in Haiti and other areas presenting improved
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27 117 efficiencies for the removal of human viral pathogens from water. Moreover, the concentration of viral
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29 118 pathogens in urban sewage and water sources in Haiti has been tested by evaluating if concentrations of
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31 119 viruses in urban sewage are similar to those in other reported studies worldwide (Bofill-mas et al. 2013),
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33 120 thus highlighting the need for developing improved CWFs that also remove water-borne viral pathogens.
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37 121 **Methods & Materials**

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40 122 Environmental screening was performed in the Metropolitan Area of Port au Prince, the capital of Haiti.
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42 123 Sampling took place in July 2011 during the rainy season. Water samples of 10 L were collected for viral
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44 124 analysis from 6 sampling points: two different sites along the main river (*Riviere Grise*) that flows through
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46 125 the cities (each by triplicate) that are potentially used at domestic level, one borehole and three different
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48 126 sites (at least in duplicate) at open canals that receive domestic wastewater and discharges directly into the
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51 127 Caribbean Sea.

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55 128 Water samples were analyzed within 6 hours after collection to determine *E. coli* concentration at the
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57 129 University of Quisqueya facilities in Port au Prince using direct plate analysis on ChromoCult© Coliform

1 130 Agar (Merck) (ISO 8199:2005). On the same day, viruses were concentrated from the 10L water samples by
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3 131 flocculation using a protocol based on organic flocculation with skimmed milk (Calgua et al. 2008). The
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5 132 resulting, neutralized floccule was kept at -20°C until shipment to Barcelona for analysis of viruses by PCR.
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7 133 Floccules of a total 10 L sample were then centrifuged and resuspended in 10 mL of phosphate buffer
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9 134 before nucleic acid extraction of 140 µL. Viral nucleic acids from concentrates were extracted using the
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11 135 QIAamp Viral RNA Mini Kit® (Qiagen, Valencia, Spain) following the manufacturer's instructions. Each
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13 136 sample was eluted in a final volume of 80 µL. Human adenoviruses (HAdV), JC polyomavirus (JCPyV) and
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15 137 norovirus (NoV) were quantified using qPCR and qRT-PCR as previously described (Jothikumar et al.
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17 138 2005; Bofill-Mas et al. 2006; Pal et al. 2006), and direct samples and their 10-fold dilutions were tested.
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19 139 Standard controls were used in all assays, including negative and positive controls (viral suspensions of
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21 140 HAdV2 and clinical samples that were positive for NoV), with external controls for inhibition using
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23 141 standard DNA (Albinana-Gimenez N et al. 2009).
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27 142 Positive samples of HAdV by qPCR were amplified by nested PCR (Allard and Albinsson 2001) and
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29 143 purified using the QIAquick PCR purification kit (Qiagen, Valencia, USA). Both strands of the purified
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31 144 DNA amplicons were sequenced with the ABI PRISM™ Dye Terminator Cycle Sequencing Ready
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33 145 Reaction kit with Ampli Taq® DNA polymerase FS (PerkinElmer, Applied Biosystems, Foster, CA, USA)
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35 146 following the manufacturer's instructions. The results were analyzed using the ABI PRISM 3730 XL
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37 147 automated sequencer (PerkinElmer, Applied Biosystems), and the sequences were compared with the
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39 148 nucleotide sequences present in GenBank using the BLAST program of NCBI (National Center for
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41 149 Biotechnology Information; www.ncbi.nlm.nih.gov/BLAST).
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46 150 The production of CWF took place in the ceramic workshop of *Josep Matés* (Fonteta, Girona, Spain).
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48 151 Several batches of small-scale-sized filters (1:2.5) were produced by following the production instructions
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50 152 from The Ceramics Manufacturing Working Group, and different compositions were tested. After analyzing
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52 153 the microbiological removal efficiencies, a real-scale-size filter was created to test the selected prototype.
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54 154 Small-scale-sized prototypes with different amended oxides and different firing atmospheres (oxidative vs
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56 155 reductive) were obtained with clay and rice husks from La Bisbal (Girona, Spain). The natural oxides that
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1 156 were selected for testing included Fe_2O_3 , Fe_3O_4 and Al_2O_3 and were added at 16% weight into the milled
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3 157 clay before forming the filter. Each clay batch was mixed with pulverized rice-husk that was screened with
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5 158 a sieve (250 μm) at 23% weight. Filters were pressed in a plaster mold, removed, stamped with a number
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7 159 and dried before firing at 950-990°C for 8–9 h in an oxidative atmosphere, except for those produced to test
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10 160 the effect of different firing atmospheres. Those filters with no added oxides and the same proportion of
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12 161 rice-husks were fired in oxidative or reductive atmospheres (two replicates per atmosphere) with three
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14 162 different origin clays from La Bisbal (non-calcareous) and two samples from Haiti: Artibonite clay (calcic
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16 163 based) and Aquin clay (non-calcareous).

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19 164 Firing in reductive atmosphere consist on firing in a traditional kiln with wood up to the desired temperature
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21 165 (950-990°C) and then to seal the kiln with the last coal inside the firing chamber. By this technique the
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23 166 oxygen consumes totally and a reduction atmosphere affects all clay components as the iron oxide from the
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25 167 clay that becomes ferrous oxide, then we can observe how within the wall, the colour is grey, not brown
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28 168 neither red.

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31 169 After firing, the filters were cooled down and immersed in water over night. The flow rate was measured
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33 170 after filling the wet filters with water, and filters with acceptable flow rates between 1–3 L/hr were
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35 171 approved for consequent analysis. No colloidal silver solution was added at any step. A modification of the
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37 172 filtering protocol was also tested by adding a 5-cm layer of crushed ceramic sitting inside the pot in the final
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39 173 prototype to increase the removal efficiency. Ceramic gravel was produced with crushed clay from broken
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41 174 filters fired in reductive atmosphere. After manual crushing, gravel was sieved with a superior 2.0 mm and
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43 175 inferior 0.5 mm sieve size. Five centimetres layer was selected as the maximum depth to keep an acceptable
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45 176 flow rate.

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50 177 Finally, two replicates of the final prototype in real scale (1:1) were performed to evaluate the long-term
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52 178 efficiencies and applicability of the best-performing prototypes. The total numbers of filter units analyzed
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54 179 are shown in Table 1.
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1 180 To characterize the chemical and mineralogical composition and texture of the raw material that was used
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3 181 and the ceramics that were obtained in this work, several assays were performed on a representative sample
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5 182 after homogenizing the material: X-ray Fluorescence analysis using a Sequential X-ray Spectrophotometer
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7 183 Phillips PW 2400, X-Ray diffraction by an automatic PANalytical X'Pert system, pore size distribution
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10 184 measurements by MIT (Mercury Intrusion Porosimeter, AutoPore IV 9500 Series, Micrometrics[®]),
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12 185 gravimetric nitrogen Brunauer–Emmett–Teller analysis using a Micromeritics ASAP 2000 Micropore
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14 186 Analyzer and Z-potential distribution analysis using Zeta Master of Malvern Instruments.

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17 187 Prototypes of the small-scale filters were prepared and tested using fresh water spiked with *E. coli*, Human
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19 188 Adenovirus 2 (HAdV2), DNA virus, and the coliphage MS2 as a representative of smaller sized ssRNA
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21 189 viruses (27 nm). Water from the tap (with turbidity <5 NTU and pH 8) was conditioned by adding sodium
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23 190 thiosulfate at 10% to inactivate free chlorine and stocks of challenge microorganisms were added to reach a
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25 191 minimum final concentration of 10⁵ IU/mL.

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29 192 To quantify the efficiencies of bacterial reduction, *E. coli* (Spanish Collection of Type Cultures, CECT 515)
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31 193 was cultured in Tryptone Soya Broth (TSB) and quantified after filtration and PBS dilution by plating on
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33 194 Trypticase Soy Agar (TSA).

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37 195 The viruses spiked were: HAdV2 and MS2 as DNA and RNA viral models, respectively. HAdV2 was
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39 196 cultured in A549 cell culture and quantified by qPCR (Bofill-Mas et al. 2006). MS2 was obtained from the
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41 197 American Type Culture Collection (ATCC 15597-B1), was multiplied by adding MS2 suspension to a log-
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43 198 phase host culture (*Salmonella* WG49, CECT 4625) and was enumerated following the double agar layer
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45 199 procedure. At the same time, Norovirus GII obtained from clinical samples was used to spike-challenge the
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47 200 water samples to add evidence for the viral removal of the selected filters.

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51 201 To assess the capability of the ceramic filter prototypes and to eliminate viruses and bacteria, the small
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53 202 filters (1:2.5) that were amended with metal oxides and filters fired in different atmospheres (reductive vs
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55 203 oxidative) were tested in a set of microbiological-removal efficiency studies. Each prototype was tested at
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57 204 least in two parallel filters. After production, the ceramic filters were saturated in mineral water overnight,
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1 205 and blank tests were performed by pouring mineral water through the filter. One liter of the challenge water
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3 206 was filtered in each test at least twice for each prototype.
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6 207 Log reduction values (LRV) were calculated by subtracting the log₁₀ of the final concentration from the
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8 208 log₁₀ of the initial concentration, and mean values and standard deviations were calculated. The Wilcoxon
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10 209 rank sum test was applied to detect significant differences in LRV between tested filters prototypes vs the
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12 210 control filters (no added oxides + oxidative firing); p-values are shown in Table 6.
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16 211 The black filter (fired in a reductive atmosphere) with an extra 5-cm layer of ceramic gravel (2-5 mm) was
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18 212 the selected prototype based on the microbiological reduction values of the previous experiments. To
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20 213 evaluate its long-term performance, the WHO-testing recommendations for HWTS (WHO 2011) were used
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22 214 with two 1:1-scale prototype replicates of fired pieces with a final diameter of 320 mm and height of 230
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24 215 mm. The performance of the selected models was monitored for over 1000 filtered liters and a daily dosing
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26 216 of 40 liters of dechlorinated, municipal tap water. Challenge waters were spiked with the selected
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28 217 microorganisms (as in previous experiments) once per week and allowed to filter for 4 hours before the
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30 218 effluent was collected to evaluate filter performance. Water quality parameters such as pH and flow rate
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32 219 were measured daily.
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37 220 **Results and Discussion**

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40 221 The results obtained in the water samples in Haiti showed high levels of human fecal contamination in the
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42 222 superficial waters of the Metropolitan Area of Port-au-Prince during July 2011 and are summarized in Table
43
44 223 2. HAdV were detected in high amounts in water from open canals that were considered to contain urban
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46 224 sewage (4.35E+04 GC/100 mL, 6/6), as well as at the two river-water-sampling sites (1.03E+02 GC/100
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48 225 mL, 4/6) and at the borehole level (3.9E+04 GC/100mL). These results are in accordance with previous
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50 226 studies, which showed the abundance of HAdV and its use as indicators of human contamination (Bofill-
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52 227 Mas et al. 2006). Important viral enteric pathogens that produce gastroenteritis were also detected: HAdV
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54 228 type 40-41, NoV GI and NoV GII, which emphasizes the importance of implementing water and sanitation
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56 229 programs that are focused on the reduction of viral infections in the population. *E. coli* mean levels were
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1 230 1.93E+04 cfu/100 mL (6/6) in the river samples and 1.74E+06 cfu/100 mL (6/6) in the sewage samples. The
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3 231 mean values of another viral indicator of human origin, JCPyV, were quantified, and the mean value in
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5 232 sewage was 3.46E+02 GC/100 mL (5/6); it was not detected in river samples, but it was found in borehole
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7 233 water. Noroviruses were quantified, presenting NoV GI mean values of 2.12E+02 GC/100 mL (2/13), and
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10 234 NoV GII mean values of 2.12E+03 GC/100 mL (2/13).

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13 235 The results obtained using the bacterial standards, viral indicators and tested pathogens are a strong
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15 236 indication of the need for using household-based water treatments and improving CWFs that reduce the
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17 237 level of viral contamination in water.

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20 238 The prototypes of the CWFs that were developed in this study and tested for microbiological efficiency are
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22 239 shown in Table 1. At the same time, the analysis of the chemical composition of the clays used to produce
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24 240 those CWFs indicates that silica is the major component, and the main difference between the three clays is
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26 241 the calcium content: Artibonite clay had the highest percentage (Table 3). After firing, all prototypes
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28 242 presented acceptable levels of water filtration capabilities and robustness. The main mineralogical
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30 243 differences between the ceramics that were fired in reducing and oxidizing atmospheres were the presence
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32 244 of magnetite or hematite, respectively, as quartz and feldspar were the dominant phases in both samples.
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34 245 Moreover, we observed that, in oxidizing conditions, the proportion of calcite (CaCO_3) in the ceramic was
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36 246 lower and the content of gehlenite ($\text{Ca}_2\text{Al}(\text{AlSi})\text{O}_7$) was increased because, in this case, the decomposition
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38 247 was more efficient. However, during reductive firing, the atmosphere seems to restrain the decomposition,
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40 248 which indicates remaining values of calcite (Table 4). Pore-size-distribution studies of the two firing
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42 249 conditions did not show significant differences between ceramics fired in different atmospheres regarding
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44 250 the pore size distribution showing larger dimensions than viral diameters (Figure 1). Nevertheless,
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46 251 significant differences were found regarding the analysis of the surface area of the ceramic filters, which
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48 252 indicated that filters fired in a reductive atmosphere had greater surface areas, with a mean value of 6.65
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50 253 m^2/g , than the filters that were fired in an oxidative atmosphere, with a mean surface area of 2.41 m^2/g
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52 254 (Table 5). Moreover, studies of Z-potential distribution of ceramics comparing both firing conditions shows
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1 255 a fraction of the crushed reductive clay being positive, suggesting a potential effect into viral absorption
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3 256 (Figure 3).
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6 257 The microbiological removal efficiency of the diverse prototypes was tested with a minimum of 2 filter
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8 258 replicates per prototype, and the results are shown in Table 6. The filter control, which was produced as a
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10 259 reference model of a non-silver-impregnated filter, presented viral and bacterial removal efficiencies of 0.57
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12 260 for MS2 and 0.68 for *E.coli*, results that are equivalent to previously reported studies (van der Laan et al.
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14 261 2014).
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18 262 The prototype ceramic filters that were amended with Fe_2O_3 , Fe_3O_4 and Al_2O_3 oxides did not significantly
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20 263 increase the efficiency of disinfection when compared to the control filters. The Log Reduction Values that
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22 264 were observed were lower than 1 logarithm for all types and for each microorganism that was tested, except
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24 265 for the filters with Al_2O_3 , which showed a slightly significant increase in *E.coli* removal of 1.34 LRV (p
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26 value 0.046). In contrast, filters that were fired in a reductive atmosphere and produced from 3 clays of
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28 267 different origins (Spain and Haiti) had greater removal efficiencies for viruses (HAdV, MS2 and NoV) and
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30 268 bacteria (*E.coli*) around 3.0 log and 2.5 log, respectively being significant compared to control filters (Table
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32 269 6). The analysis of the log removal efficiencies among reductive atmosphere filters were no significant
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34 270 ($p>0.05$). These filters achieved the performance requirements for of the HWTS protective technologies that
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36 271 were recently established by the WHO (WHO 2011).
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41 272 Filters fired in a reductive atmosphere with an extra layer of ceramic gravel were selected to test long-term
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43 273 performance in real size scale and showed consistent removal values after 1000 liters (Figure 2). During the
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45 274 long-term study, logarithm reduction values were between 2.5-4 LRV for the tested viruses (HAdV and
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47 275 MS2), but higher variations occurred for *E.coli*, with a range of 1.5-4 LRV. Flow rates were recorded at 1.5-
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49 276 3 liters/hour and were stable throughout the experiment, and the effluents had a slightly increased pH, from
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51 277 7 in the input water up to 8 in the output water, as previously recorded (data not shown) (Halem 2006).
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55 278 The new model of a Ceramic Water Filter made by firing in a reductive atmosphere and without colloidal
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57 279 silver that have been proposed in this paper showed removal efficiencies for viruses and bacteria that were
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1 280 significantly higher than the current standards and what was previously reported in the literature.
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3 281 Furthermore, they reach the WHO targets for HWTS technologies. Black ceramics, which are clay fired in a
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5 282 reductive atmosphere, seems to increment the internal specific surface area and the Z-potential becoming
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7 283 the most feasible model for achieving advantageous results in viral removal of the proposed models. This
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10 284 model seems to be a low-cost option whose production can be easily adapted for a wide-spread model of
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12 285 CWF to increase microbiological effectiveness without increasing costs.
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15 286 **Conclusions**

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17 287 High levels of waterborne viruses are present in superficial waters of Port-au-Prince, Haiti, highlighting the
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19 288 importance to improve household water technologies for viral removal efficiencies. A new prototype of
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21 289 CWF fired in reductive atmosphere, a variation that maintains the product non-expensive, improves viral
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23 290 removal efficiencies up to 3 logs, accomplishing the WHO requirements for HWTS technologies. Long-
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25 291 term assays with this new prototype show consistent removal values after 1000 liters filtered. Ceramics
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27 292 fired in reductive atmosphere show greater surface area and higher z-potential the most feasible explanation
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29 293 for viral disinfection properties of this new model.
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34 294 This new improved prototype will be easily implemented in CWF factories as firing in reductive atmosphere
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36 295 will just require a specific kiln that can be built with local materials (traditional bricks, sand and concrete) or
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38 296 even current kiln models can be accommodated to fire in reductive conditions. Moreover, the fact that
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40 297 colloidal silver can be replaced by a layer of ceramic gravel will also reduce environmental and toxicity
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42 298 hazards as well as economic cost (around 1 euro per filter). At the same time, the addition of this layer made
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44 299 of ceramic gravel will entail a step of maintenance by users because regular cleaning of gravel will be
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46 300 needed. However, this modification will also improve the microbial performance of the filter and highlights
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48 301 the importance of performing hygiene promotion campaigns alongside the implementation of this product
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50 302 towards users.
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54 303 **Acknowledgements**

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1 304 Intermon Oxfam and University of Barcelona (International cooperation and development grant, 2011)
2
3 305 funded this study. We acknowledge Josep Matés for his tremendous contribution in the production of
4
5 306 ceramic prototypes for this study. We thank the collaboration Montserrat Español from the Department of
6
7 307 Materials Science and Metallurgical Engineering (CMEM) UPC and the Physicochemical Department of
8
9 308 Pharmacy Faculty, UB, participating in the study. During the development of this study, Marta Rusiñol was
10
11 309 a fellow of the Catalan Government “AGAUR” (FI-DGR).
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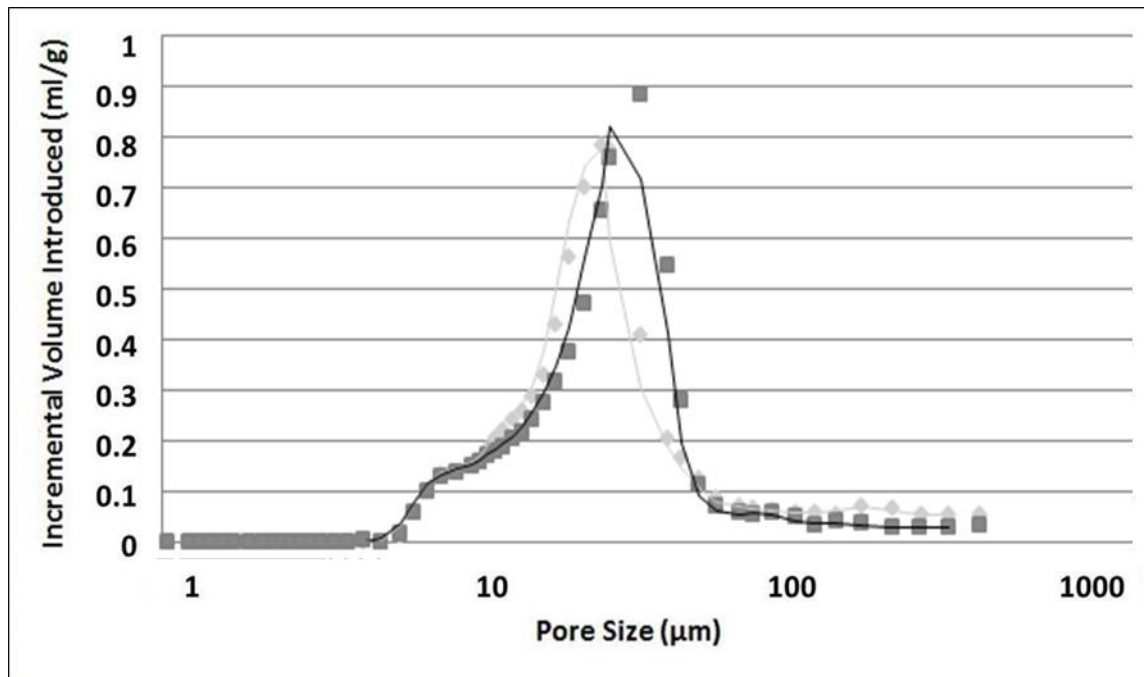


Figure 1 | Pore size distribution of ceramics that were fired in oxidative (squares) and reductive atmospheres (rhombus).

Figure 2

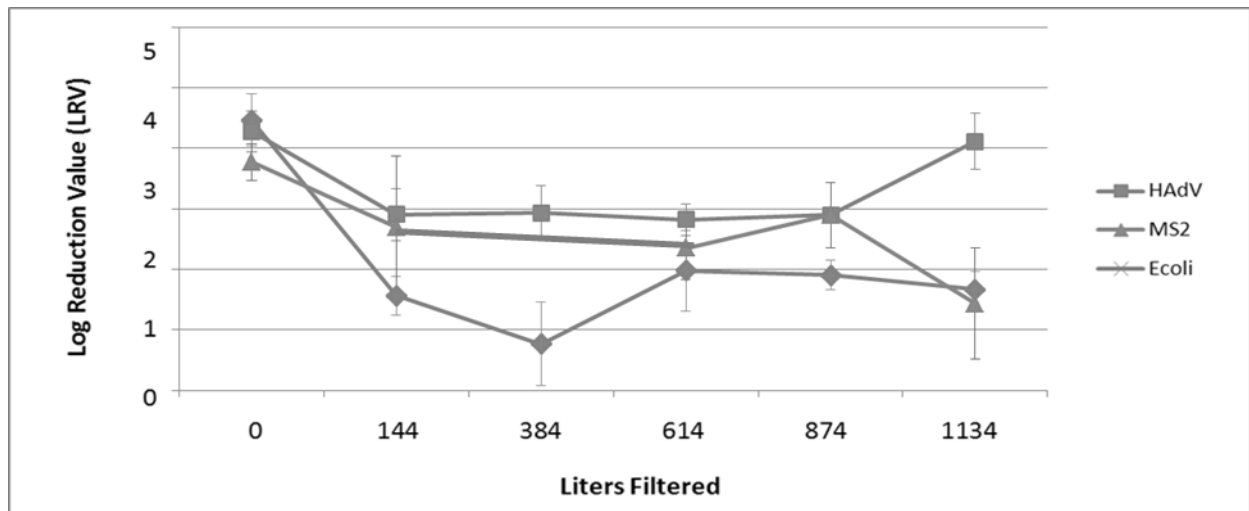


Figure 2 | Monitoring of long-term microbiological effectiveness of the selected CWF (reductive atmosphere + 5-cm ceramic gravel layer).

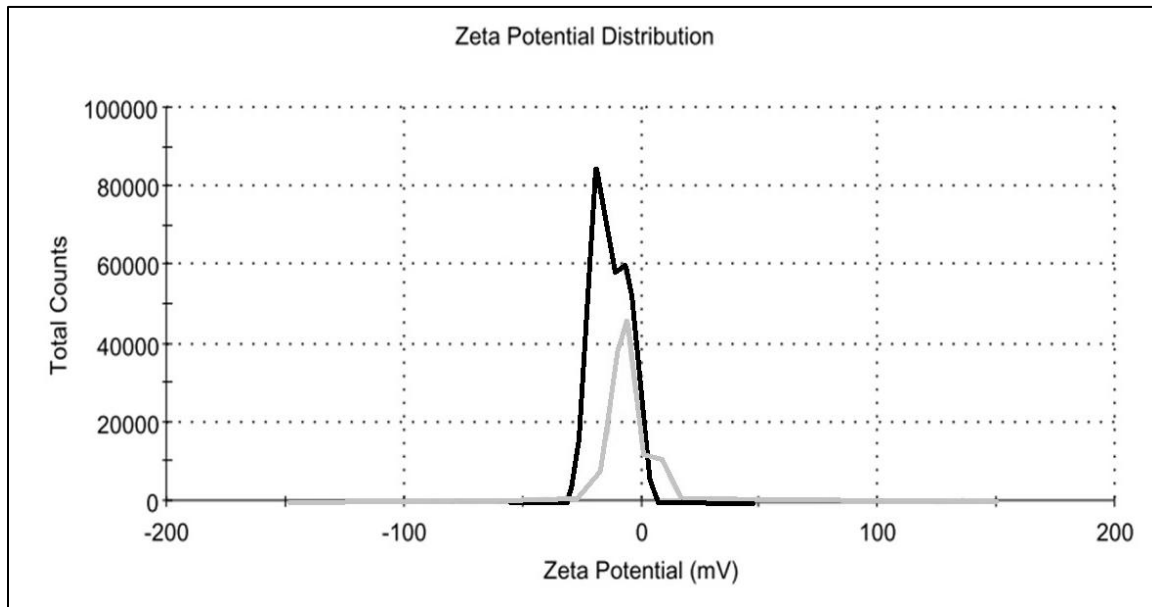


Figure 3 | Zeta Potential Distribution of Clay from la Bisbal fired in Oxidative Atmosphere (black line) and Clay fired in Reductive Atmosphere (grey line).

Table 1. Prototypes of the produced Ceramic Water Filters

Ref	Ceramic additives	Clay origin	Firing atmosphere	Scale	Layer of ceramic gravel (cm)	Replicates
1	None	Bisbal, Spain	oxidative	1:2.5	-	4
2	+ 16% Fe ₂ O ₃	Bisbal, Spain	oxidative	1:2.5	-	2
3	+ 16% Fe ₃ O ₄	Bisbal, Spain	oxidative	1:2.5	-	2
4	+ 16% Al ₂ O ₃	Bisbal, Spain	oxidative	1:2.5	-	2
5	None	Bisbal, Spain	reductive	1:2.5	-	4
6	None	Bisbal, Spain	reductive	1:2.5	5	2
7	None	Artibonite, Haiti	reductive	1:2.5	-	2
8	None	Aquin, Haiti	reductive	1:2.5	-	2
9	None	Bisbal, Spain	reductive	1:1	5	2

Table 2. Microbiological contamination in superficial waters of Port au Prince

Location	Type	N	<i>E.coli</i>	HAdV	JCPyV	NoV GII	NoV GI
			cfu/100 ml (positive/total)	GC/100 ml (positive/total)			
Cité du Soleil	Canal	1	9.00E+05	1.61E+03	3.25E+01	ND	ND
Bois Chene	Canal	1	3.00E+06	3.91E+03	2.64E+01	3.63E+03	ND
		2	5.00E+06	6.15E+03	1.36E+03	ND	ND
		3	1.70E+05	1.66E+05	ND	ND	ND
Canal Brea, Martisand	Canal	1	1.20E+06	3.31E+04	3.11E+01	ND	ND
		2	1.40E+05	5.03E+04	2.81E+02	ND	2.55E+02
Mean Value Canal			1.74E+06	4.35E+04	3.46E+02	3.63E+03	2.55E+02
			(6/6)	(6/6)	(5/6)	(1/6)	(1/6)
Riviere Grise, Croix de la Mission	River	1	1.70E+03	ND	ND	ND	1.68E+02
		2	4.00E+04	7.77E+01	ND	ND	ND
		3	4.00E+03	ND	ND	ND	ND
Riviere Grise, Pont sur la route #9	River	1	1.00E+04	1.17E+02	ND	ND	ND
		2	2.00E+04	7.49E+01	ND	ND	ND
		3	4.00E+04	1.41E+02	ND	6.01E+02	ND
Mean Value River			1.93E+04	1.03E+02	-	6.01E+02	1.68E+02
			(6/6)	(4/6)	(0/6)	(1/6)	(1/6)
Tabarre	Borehole	1	2.60E+01	3.94E+04	5.17E+01	ND	ND

ND: Not detected

Table 3. Chemical analysis of clays used to make CWF

Chemical composition	Weight (%)		
	LaBisbal, Girona, Spain	Artibonite, Haiti	Aquin, Haiti
SiO ₂	62.87	43.77	66.60
Al ₂ O ₃	15.88	12.86	17.32
Fe ₂ O ₃	5.56	6.96	3.12
MnO	0.07	0.07	0.02
MgO	1.20	3.65	0.45
CaO	1.53	11.94	0.33
Na ₂ O	0.41	1.16	0.66
K ₂ O	3.17	1.57	0.19
TiO ₂	0.78	0.75	0.24
P ₂ O ₅	0.13	0.14	0.04

Table 4. Mineralogical analysis of ceramics filters fired in different atmospheres in percentages (%)

Mineral (%)	Artibonite		La Bisbal	
	reductive	oxidative	reductive	oxidative
Quartz	31	23	51	47
Sericite-heated	-	-	44	40
Diopside	11	11	-	-
Calcite	4	2	-	-
Lime	1	0.5	-	-
Gehlenite	4	6	-	-
K feldspar	12	13	3	5
Plagioclase	35	43	-	7
Hematite	-	2	-	1
Magnetite	1	-	2	-

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Table 5. Specific surface areas of Artibonite and La Bisbal filters that were fired in different atmospheres.

Firing atmosphere	BET (m ² /g)	
	Artibonite	La Bisbal
Reductive	6.8713 (± 0.0475)	6.4371 (± 0.0120)
Oxidative	2.6185 (± 0.0281)	2.2036 (± 0.0152)

Table 6. Microbiological effectiveness of different Ceramic Water Filter prototypes in Log₁₀ Reduction Values and statistical significance levels from Wilcoxon rank sum test.

Ref	Filter	Microorganism	Log Reduction Value				
			Mean	n ¹	Std. Deviation	P value	
1	CWF-Bisbal-oxidative	HAdV	0.62	18	0.43	ref	
		MS2	0.57	8	1.04		
		NoV	0.96	2	0.32		
		E. coli	0.68	12	0.62		
2	CWF-Bisbal-oxidative + Fe ₂ O ₃	HAdV	0.67	10	0.47	0.74	
		MS2	0.54	2	0.11	0.27	
		E. coli	0.59	8	0.69	0.87	
3	CWF-Bisbal-oxidative + Fe ₃ O ₄	HAdV	0.57	9	0.37	0.72	
		MS2	0.25	2	0.01	0.26	
		E. coli	0.53	8	0.82	0.46	
4	CWF-Bisbal-oxidative + Al ₂ O ₃	HAdV	0.85	9	0.37	0.21	
		MS2	0.26	2	0.03	0.27	
		E. coli	1.34	7	0.55	0.046	*
5	CWF-Bisbal-reductive	HAdV	3.54	14	0.73	<0.001	*
		MS2	2.33	8	0.61	0.02	*
		E. coli	2.32	8	0.85	0.005	*
6	CWF-Bisbal-reductive + gravel	HAdV	4.15	2	0.12	0.008	*
		MS2	2.97	2	0.11	0.024	*
		NoV	3.56	2	0.23	0.667	
		E. coli	2.90	2	0.28	0.035	*
7	CWF-Artibonite-reductive	HAdV	3.36	2	0.62	0.008	*
		MS2	2.98	2	0.12	0.024	*
		E. coli	2.37	2	0.47	0.035	*
8	CWF-Aquin-reductive	HAdV	2.86	2	0.24	0.008	*
		MS2	1.27	2	0.02	0.027	*
		E. coli	2.40	2	0.43	0.035	*

¹ n = number of assays

* Significance levels of 5% or less (p≤0.05)