Journal of Water & Health



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Journal of Water and Health Vol 19 No 5, 750 doi: 10.2166/wh.2021.052

Performance evaluation of ceramic pot filters combined with adsorption processes for the removal of heavy metals and phenolic compounds

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ABSTRACT

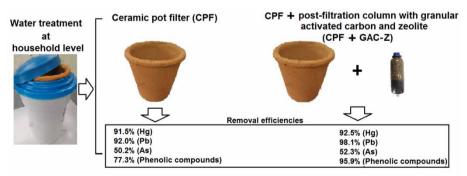
It has been demonstrated that the ceramic pot filters (CPFs) with impregnated colloidal silver are efficient for the removal of turbidity and pathogens for household water treatment. This investigation evaluated the efficiency of two filter models for the removal of chemical contaminants (Hg, Pb, As and phenolic compounds) during 175 days. The first model is a traditional CPF impregnated with colloidal silver and the second consists of the ceramic silver-impregnated pot plus a post-filtration column with granular activated carbon and zeolite (CPF + GAC-Z). The results of the CPF showed average efficiencies of 91.5% (Hg), 92% (Pb), 50.2% (As) and 78.7% (phenols). The CPF + GAC-Z showed similar efficiencies for the removal of heavy metals (92.5% Hg, 98.1% Pb and 52.3% As) and a considerably higher efficiency for the removal of phenols (96.4%). The As concentration of the filtered water in both systems was higher than the regulatory limit. The ceramic pot was responsible for the highest removal of chemical compounds. It can be concluded that the traditional CPF is a viable option for water supply treatment at the household level for the removal of chemical contaminants. The efficiency of this filter can be improved with the post-filtration column mainly for the removal of organic constituents.

Key words: ceramic pot filter, chemical risk, heavy metals, household water treatment, water supply

HIGHLIGHTS

- The ceramic pot filter (CPF) is a viable household technology for chemical contaminants.
- The combined use of the ceramic pot with granular activated carbon and zeolite improves the removal of organic contaminants.
- The CPFs evaluated in this study reached efficiencies higher than 90% for mercury and lead removal.
- The efficiencies for arsenic and phenols were higher than 50 and 79%, respectively.

GRAPHICAL ABSTRACT



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INTRODUCTION

Although microbiological contamination remains the largest cause of water-related morbidity and mortality globally, chemicals in water supplies may also cause diseases, and evidence of the effects on human health is either limited or lacking for many of them (Villanueva *et al.* 2014). Exposure to these chemicals typically occurs at low concentrations and through multiple routes.

The list of chemical contaminants continues to increase, and the impact on human health has not been evaluated in most cases. This is due to a lack of reference data in many countries and, often, also due to the low sensitivity of the available analytical techniques (Villanueva *et al.* 2014).

The chemical contamination of water supplies is strongly influenced by anthropogenic activities that include agriculture, mining and industry involving point sources, infiltration or runoff. The chemical exposure (e.g. heavy metals) through consumption of drinking water is almost unavoidable, people in small and rural communities being the most vulnerable (Chowdhury *et al.* 2016).

Previous studies carried out in Dalian, China, show the health risk caused by the presence of chemical contaminants, such as hexachlorobenzene, and heavy metals like arsenic, as well as pesticides in water sources and sediments, which were the main contributors to human carcinogenic risks (Dong *et al.* 2020). Similar studies (Evans *et al.* 2019) found that the cumulative risk analysis of contaminant occurrence in drinking water in the United States indicates that over 100,000 lifetime cancer cases could be ascribed to carcinogenic chemicals in tap water. The larger part of this risk is attributable to the presence of arsenic, disinfection by-products and radioactive contaminants.

Among the contaminants of recognized importance are heavy metals such as arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn). All of these metals are frequently present in industrial products and are widely recognized as a potential hazard to human health and ecosystems when their exposure concentrations exceed critical levels (WHO 2017). Hg, Pb and As were selected as the targeted heavy metals in this study because they have been increasingly recognized as major threats to Colombian water resources quality with origins in mining and agricultural activities (Marrugo-Negrete *et al.* 2008; Alonso *et al.* 2014; Torrance *et al.* 2021).

In many cases, the exposure to high concentrations of heavy metals affects vulnerable populations like children, the elderly and immune-deficient patients. In many Latin American countries (Argentina, Bolivia, Brazil, Chile, Colombia, Cuba, Ecuador, El Salvador, Guatemala, Honduras, Mexico, Nicaragua, Peru and Uruguay), extremely high As levels of anthropogenic origin were detected in nearly all possible drinking water sources, including springs, lakes, rivers and groundwater (Khan *et al.* 2020).

Phenol and its derivatives are also considered a threat to public health and aquatic ecosystems. Phenolic compounds, like 2-chlorophenol, 2,6-dichlorophenol, 2,4,5-trichlorophenol, 2,4-dimethylphenol, 4-nitrophenol and p-cresol, have been detected worldwide in water supply sources at concentration levels not deemed suitable for drinking purposes (Nthunya *et al.* 2019). These phenolic compounds can have a natural origin (through the decomposition of dead plants and animals in the water), as well as a result of anthropogenic activities (through industrial, domestic, agricultural and municipal activities). The main industrial uses include paints, fertilizers, surfactants, explosives, textiles, rubbers, plastics, curing agents and antioxidants, besides being constituents of some pesticides and insecticides (Anku *et al.* 2017).

Improving water quality at the tap and investing in measures to protect water sources represent opportunities for protecting public health and decreasing potential disease incidence due to environmental pollution (Evans *et al.* 2019). For small/rural communities and individuals, point-of-use treatments are an alternative to ensure safe drinking water (Dong *et al.* 2020).

The World Health Organization (WHO) includes household treatment technologies as a part of its strategies within its integral frame for risk management (WHO 2017) in order to guarantee water supply quality from a chemical and bacteriological point of view. To reduce the microbiological risk, it is recommended to use technologies such as chemical disinfection, membranes, porous ceramic filters, granular filters, solar or UV disinfection, thermic technologies, coagulation, precipitation with sedimentation and combined treatments. These technologies are considered to control the chemical risk posed by priority contaminants, such as fluorides, arsenic and nitrates through anionic and cationic exchange, activated carbon and lowpressure units of inverse osmosis.

The ceramic pot filter (CPF) is a household filter technology operated by gravity flow. The CPFs are manufactured with clay mixed with sawdust or rice husk and impregnated with colloidal silver. The porosity of the filter is created as the sawdust or rice husks are burned and leave behind pore spaces (Brown & Sobsey 2010). These filters have been successfully used to remove turbidity and pathogen microorganisms (Mwabi *et al.* 2011; WHO 2017; Akosile *et al.* 2020). However, information related to the efficiency of these systems for the removal of heavy metals and organic pollutants in the ceramic pot remains

scarce. A recent study at the laboratory scale demonstrated that the use of CPFs impregnated with colloidal silver reached removal efficiencies of 57.3% chromium(VI), 69.1% methyl orange and 100% of *E. coli*. In addition, the same study recommended that an up-scaled version of the filter should be useful in treating water in low-income communities, even under natural disaster conditions (Chaukura *et al.* 2020a). According to Chaukura *et al.* (2020b), further research should focus on functionalizing the biochar layer in CPFs and investigate the removal of toxic metals and specific organic pollutants.

The CPF systems have the potential to remove chemical constituents by adsorption in the pot (Ludeña & Tinoco 2010; Guerra-Alarcón & Hiyagon-Arroyo 2012) or by the combination of filtration systems that use adsorbent materials like zeolite, iron, diatomite and biochar (Akhigbe *et al.* 2016; Ekpunobi *et al.* 2019; Chaukura *et al.* 2020b).

The main materials of a CPF are clay and a combustible burnout material, such as milled sawdust or rice husks, which are characterized by a high concentration of oxides and iron hydroxides, as well as carbon with a high pollutant adsorption capacity (Chaukura *et al.* 2020b). Adsorption is a mechanism commonly applied for the removal of heavy metals and is more attractive for household-level water treatment in view of it being an effective technique and its low cost (Prathna *et al.* 2018). Previous studies have demonstrated that few low-cost adsorbents, when filled into a column, reduced As, Cu and Pb significantly. Therefore, it is a challenge to improve the use of low-cost adsorbents focusing on their in-house applications (Chowdhury *et al.* 2016).

The aim of this study was to evaluate CPFs made with a combination of clay and sawdust on their efficiency in the removal of heavy metals (Hg, Pb and As) and phenolic compounds. The evaluated filters included the traditional model which consisted of a ceramic silver-impregnated pot and a plastic storage container (CPF), as well as another model that included the same constituents along with a post-filtration column to improve the final water quality. The column contained granular activated carbon (GAC) and zeolite (CPF + GAC-Z). The GAC was used by its capacity for the removal of organic chemicals, and the zeolite was used by its efficiency as an ionic exchanger that could improve the heavy metal removal (WHO 2017).

These types of household filters are being used in Colombia rural areas affected by low water supply. Previous studies have demonstrated that these systems produce safe drinking water removing microbiological contaminants like *Escherichia coli* and *Salmonella* spp. (Rivera-Sánchez *et al.* 2020).

MATERIALS AND METHODS

The research was undertaken at the Universidad Santiago de Cali, Colombia. The average environmental temperature during their evaluation was 21 °C \pm 1.0. The filters were new and are made by the Colombian company EKOFIL LTDA which has a quality certification for the production processes. The traditional model, called CPF, included a ceramic pot impregnated with colloidal silver and a 30 L plastic storage bucket. The firing temperature of the ceramic pot was 900 °C.

The colloidal silver application was made by soaking the ceramic pot into a 500 ppm solution. The second one, called CPF + GAC-Z, had an additional post-filtration column with GAC and zeolite (see Figure 1). The adsorption column was



Figure 1 | Description of CPFs used in the study.

LD (mg/L)

a 260 mL plastic bottle with an inlet and outlet adaptor. The column was filled with proportion 1:3 of carbon and zeolite. Both ceramic pots were operated at a filtration rate of around 1.8 L/h that was guaranteed by its cleaning every 15 days. The filters were cleaned with clean water and rubbing the wall with a soft bristle brush (Pérez-Vidal *et al.* 2020). The pressure loss in the filtration column was not measured.

The water level in the storage bucket was maintained at around 10 cm, and the post-filtration column was never submerged in the filtered water. The research was carried out in two steps. First, the spiked water was prepared. Secondly, the filtration systems were operated in batch mode, and the tap was opened daily to remove the filtered water.

The experimental design was unifactorially differentiated by the chemical constituents which were evaluated (mercury, lead, arsenic and phenolic compounds). Four filters were evaluated (two CPF and two CPF + GAC-Z) over a period of 175 days filtrating spiked water. Additionally, two controls were implemented filtrating tap water to evaluate the contribution of the evaluated contaminants by the ceramic pot and the filtration column.

Preparation of spiked water

For the preparation of the spiked water, tap water was dechlorinated with analytical grade sodium thiosulfate (Merck: 106512) to reach a maximum concentration of 20 mg/L. Following the General Test Water recommended by the *Guide Standard and Protocol for Testing Microbiological Water Purifiers* of United States Environmental Protection Agency – EPA, the turbidity was adjusted to a value of 5 NTU and 100 mg/L of total dissolved solids (TDS). The turbidity was adjusted by adding a 0.02 g/L of kaolin and the TDS concentration by the addition of analytical grade NaCl (Merck: 106404) to obtain a 0.12 g/L concentration.

The kaolin and NaCl doses were determined by means of dose vs. concentration curves that were validated with the final turbidity (Nephelometric Standard Method 2130B) and the TDS values (difference between total solids, Standard Method SM2540B, and total suspended solids, Standard Method SM2540D). Besides, a correlation curve between the conductivity and TDS was made in order to establish the average conductivity value correlated with a TDS of around 100 mg/L.

The daily volume of prepared spiked water was 65 L. The chemical substances were added using standard solutions of 1,000 mg/L of Hg, Pb and As (Merck: 170226, 119776 and 170238) and analytical grade 2,4,6-thrichlorophenol (98%, Merck[®]: 818,469). The doses of the chemical substances are presented in Table 1. The value range of the evaluated doses was defined by taking into account three conditions: (i) that it be higher than the regulatory range, (ii) that it be similar to the concentrations reported in contaminated waters and/or (iii) that it be higher than the limit of detection (LD) of the laboratory analytical techniques.

During the first 30 days of the study, the filters were operated with the lowest doses (phase 1) and the effluent concentrations under the LD of the analytical techniques. During phase 2, the systems were evaluated with higher doses to obtain measurable efficiencies. It should be noted that although it was intended to fix the chemical substance concentrations

		Reported value in		
Applied dose range (mg/L)	Regulatory value (mg/L)	contaminated waters (mg/L)	mg/L	Analytical method
Phase 1: 0.01 Phase 2: 0.04–0.1	0.006 ^a	0.013-0.251 ^b	>0.001	SM3112B
Phase 1: 0.08–0.1 Phase 2: 0.21–0.3	0.01 ^a	0.02-0.43 ^{c,d}	>0.01	SM3111B
Phase 1: 0.01–0.1 Phase 2: 0.3–1.1	0.01 ^a	0.014-0.255 ^{c,d,e}	>0.01	SM3114C
Phase 1: 0.92–1.7 Phase 2: 1.8–2.9	0.2 ^a	0.480-2.90 ^f	>0.002	EPA9065 (phenolic compounds)
	Phase 1: 0.01 Phase 2: 0.04–0.1 Phase 2: 0.21–0.3 Phase 1: 0.01–0.1 Phase 2: 0.3–1.1 Phase 1: 0.92–1.7	Phase 1: 0.01 0.006 ^a Phase 2: 0.04-0.1 0.01 ^a Phase 1: 0.08-0.1 0.01 ^a Phase 2: 0.21-0.3 0.01 ^a Phase 1: 0.01-0.1 0.01 ^a Phase 1: 0.92-1.7 0.2 ^a	Phase 1: 0.01 0.006 ^a 0.013-0.251 ^b Phase 2: 0.04-0.1 0.01 ^a 0.02-0.43 ^{c,d} Phase 1: 0.01-0.1 0.01 ^a 0.014-0.255 ^{c,d,e} Phase 2: 0.3-1.1 0.2 ^a 0.480-2.90 ^f	Applied dose range (mg/L) Regulatory value (mg/L) contaminated waters (mg/L) mg/L Phase 1: 0.01 0.006 ^a 0.013–0.251 ^b >0.001 Phase 2: 0.04–0.1 0.01 ^a 0.02–0.43 ^{c,d} >0.01 Phase 1: 0.08–0.1 0.01 ^a 0.02–0.43 ^{c,d} >0.01 Phase 1: 0.01–0.1 0.01 ^a 0.014–0.255 ^{c,d,e} >0.01 Phase 1: 0.01–0.1 0.01 ^a 0.480–2.90 ^f >0.002

Table 1 | Description of the evaluated chemical substances, applied doses and criteria for the doses' selected range

LD, limit of detection; SM, standard method. ^aWHO (2017). ^bSee Shoham-Frider *et al.* (2020).

^cSee Singh et al. (2020).

^eSee Alonso *et al.* (2014).

^fSee Nthunya et al. (2019) and Tosic et al. (2019).

dSee Torrance et al. (2021).

in phase 1 (Hg: 0.01 mg/L; Pb: 0.1 mg/L; As: 0.1 mg/L and phenolic compounds: 1.0 m/L) and in phase 2 (Hg: 0.1 mg/L; Pb: 0.3 mg/L; As: 0.3 mg/L and phenolic compounds: 2.0 mg/L), there was some variability due to the daily prepared volume of spiked water. Despite the variation in the chemical substance concentrations, the spiked water sample was filtered and analyzed.

Operation and performance of the filtration systems

The filtration systems were operated by adding 7.5 L of spiked water per filter, twice a day. This volume corresponds to the maximum capacity of the ceramic pot as well as to the daily minimum amount of water for drinking and food preparation per person, as recommended by the WHO. The statistical comparison of the performance of the systems was made by using boxplot and tests of variance: ANOVA and POSTANOVA using Tukey's honestly significant difference method. The statistical analysis was made with the R-Project free software. The daily values between the duplicates were averaged. Table 2 shows variables evaluated during the operation systems.

RESULTS AND DISCUSSION

Characteristics of the spiked water

Table 3 shows the average variation of the spiked water quality during the study. The conductivity was used as an indirect parameter of the TDS concentration taking into consideration that an average value of conductivity of $200 \pm 18.7 \,\mu$ s/cm is related to a TDS concentration of $112 \pm 28 \,$ mg/L.

The pH during the study showed an average near to the neutral value. The conductivity and the concentration of heavy metals were between the ranges previously defined in the methodology, with the exception of arsenic and phenolic compounds, which reported the maximum values of 1.1 and 2.9 mg/L, respectively. Although these values were higher than those initially established (see Table 1), there are reports of arsenic concentrations in water supply superseding 2 mg/L, a

Variable	Units	Frequency	Methods/equipment
рН	Unitless	Daily	$Electrometric \textbf{-4500-} H + B^a/pHmeter \ F20 \ Five EasyTM$
Turbidity	NTU	Daily	Nephelometric-2130B ^a /Turbidimeter HACH 2100N
Conductivity	μS/cm	Daily	Laboratory-2520B ^a /VWRS conductivity meter 2052
Mercury	mg/L	Biweekly	SM3112B; SM3111B; SM3114C ^a /atomic absorption
Lead	mg/L		spectrophotometer VARIAN FS240. Wavelengths: 253.7 nm (mercury), 217.5 nm (lead) and 197.2 nm (arsenic).
Arsenic	mg/L		
Phenolic compounds	mg/L	Biweekly	EPA method 9,065/Thermo Genesis 30

Table 2 | Evaluated variables during the study

^aSee APHA (2012).

 Table 3 | Physicochemical characteristics of the spiked water during the study

Variable	n	Unit	Average	Standard deviation	Minimum	Maximum
pH	95	Unitless	6.9 ^a	-	6.20	8.80
Conductivity	100	μs/cm	191.5	88.1	56.0	450.0
Turbidity	95	NTU	3.1	1.5	1.10	8.90
Mercury	12	mg/L	0.048	0.03	0.01	0.10
Lead	12	mg/L	0.227	0.09	0.08	0.30
Arsenic	12	mg/L	0.307	0.32	0.01	1.10
Phenolic compounds	12	mg/L	1.878	0.49	0.92	2.90

^aGeometric mean.

value that is 200 times greater than the value limit recommended by the WHO (Kumar *et al.* 2019), and for phenolic compounds, concentrations up to 2.9 mg/L can be reached in seawater (Tosic *et al.* 2019).

The final average turbidity value was 3.1 NTU, less than 5 NTU, the value that was initially established in the methodology. It is important to note that the turbidity was in the range value reported by other studies (Brown & Sobsey 2010).

Operation of the filtration systems

Based on the obtained average measured values during the evaluation of the CPF and CPF + GAC-Z systems, it was observed that the pH was neutral and similar in both effluents with medians of 7.1 and 7.0, respectively. Other studies have shown that in this pH value occurs the highest phenolic compound removal by clay adsorption (Kaleta 2006) but neutral pH values can limit arsenic removal (Jadhav *et al.* 2015).

The conductivity of the filtered water had mean values of $205 \pm 94 \,\mu$ s/cm for the CPF and $216 \pm 93 \,\mu$ s/cm for the CPF + GAC-Z, indicating indirectly that both systems have low average TDS removal efficiencies of 22.5 ± 19.2 and $22.3 \pm 16.2\%$, respectively. This is similar to the results of previous studies that used traditional CPFs (Chaukura *et al.* 2020b). However, studies with modified CPFs that use mixed material like diatom and biochar showed TDS removal efficiencies between 46 and 64% (Ekpunobi *et al.* 2019; Chaukura *et al.* 2020b). Figure 2 shows the results of the heavy metal (Hg, Pb and As) removal for the CPF and CPF + GAC-Z compared with the raw water (spiked water).

The global analysis of the results considering the median spiked and filtered water concentrations shows that the mercury removal efficiencies for both systems were similar with values of 91.5% for the CPF and 92.5% for the CPF + GAC-Z. The

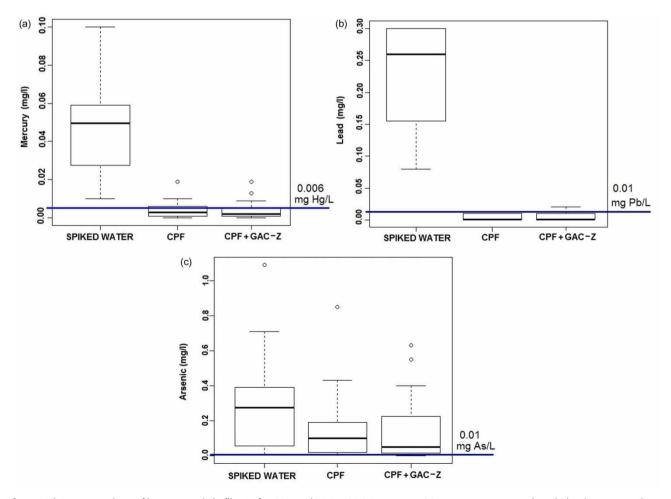


Figure 2 | Concentrations of heavy metals in filtrate for CPF and CPF + GAC-Z systems: (a) mercury concentration, (b) lead concentration and (c) arsenic concentration. The horizontal line corresponds to the limit recommended by WHO (2017).

average effluent mercury concentration of the systems was 0.004 mg Hg/L, a lower value than the recommendation by the WHO (<0.006 mg/L).

Regarding lead removal, the efficiencies were 92.1% for the CPF and 98.1% for the CPF + GAC-Z. This result is an evidence of the influence of the post-filtration column on Pb removal efficiency. The use of activated carbon can reach an removal efficiency of Pb larger than 80% (Chowdhury *et al.* 2016). The effluent average lead concentration for both systems was around 0.004 mg Pb/L, a value lower than the regulatory value established by the WHO and Colombian regulation (0.01 mg/L).

The average arsenic removal efficiency was lower compared with Hg and Pb. The efficiencies were 50.2% for the CPF and 52.3% for the CPF + GAC-Z system. The average arsenic effluent concentration for both systems was 0.15 mg As/L. This is a higher value than the prescribed value for safe water supplies (0.01 mg/L) (WHO 2017).

Figure 3 shows a longitudinal data analysis illustrating the variations of the concentrations of Hg, Pb and As during the study in the spiked water and the filtered water as well the removal efficiency for both filtration systems. In the same figure, it can be seen that the systems in phase 1 operated with a spiked water with lower concentrations than in phase 2. The blue dotted line corresponds with the regulatory limit.

It was observed that both systems could reach a higher removal efficiency for Hg and Pb than for As in both phases. Furthermore, the filtered water had concentrations lower than the regulatory value for Hg and Pb. Conversely, in both phases,

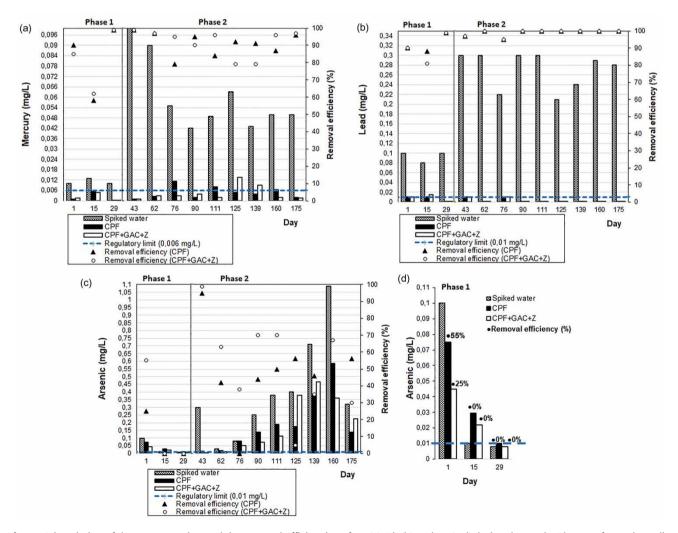


Figure 3 | Variation of the concentration and the removal efficiencies of Hg (a), Pb (b) and As (c,d) during the study. Please refer to the online version of this paper to see this figure in colour: doi:10.2166/wh.2021.052.

the As concentration of the filtered water was higher than the regulatory limit, showing potential risk of As in the treated water by CPF and CPF + GAC + Z. It is important to mention that the removal efficiencies were higher when the spiked water heavy metal concentrations were higher. This was more evident for As in phase 1 (see Figure 3(d)). However, both filtration systems showed low As removal efficiency.

The global average removal efficiency analysis based on each data shows a light variation for Hg (CPF: 88.9%; CPF + GAC + Z: 89.5%), Pb (CPF: 97.4%; CPF + GAC + Z: 96.8%) and a lower removal efficiency for As (CPF: 38.3%; CPF + GAC + Z: 44.3%). Also, it is important to mention that both systems reached higher efficiencies for Hg (99%) and Pb (100%). Although it was not possible to evaluate the saturation of the filtration columns, there was evidence that their efficiency was not reduced during the study.

In phase 1, the CPF filter reached a Hg and Pb removal efficiency between 90 and 99% and between 0 and 25% for As. The CPF + GAC + Z filter showed a Hg and Pb removal efficiency between 62 and 99% and between 81 and 99%, respectively, and between 0 and 50% for As. In phase 2, the CPF filter reached a Hg and Pb removal efficiency between 79 and 99% and for As between 42 and 56%. The Hg, Pb and As removal efficiencies in the CPF + GAC + Z filter were between 79 and 99, 95 and 100, and 30 and 70% respectively.

The ANOVA test showed no significant differences in the removal efficiencies of Hg (df = 3, F = 0.192, p = 0.902), Pb (df = 3, F = 0.085, p = 0.968) and As (df = 3, F = 0.686, p = 0.566) for both systems. The measurement of the variables in the filtered water of the negative control was less than the LD for all determinations. These results demonstrate that in the CPF + GAC-Z system, the ceramic pot is responsible for the largest contribution to the heavy metal removal efficiency. The results of this study are consistent with other experiment-based studies (Ludeña & Tinoco 2010; Guerra-Alarcón & Hiyagon-Arroyo 2012), in which removal efficiencies between 50 and 100% for mercury and 98.6% for lead were obtained.

The removal efficiencies for these metals in the filtration systems is attributed to the adsorption capacity of the clay in the pot, which is related to its texture characteristics (specific surface and porosity) and two main removal processes: absorption (capillarity retention) and adsorption (chemical interaction between the adsorbent, in this case between the clay and the absorbed substance) (Guerra-Alarcón & Hiyagon-Arroyo 2012). Adsorption is the main removal mechanism attributed to the ceramic pot. Also, water pH is a factor impacting removal efficiency (Chaukura *et al.* 2020a).

The high quantity of nanoparticles like iron oxides and hydroxides in the ceramic pot, which have a high adsorption capacity of heavy metals, promotes the removal of these contaminants (Prathna *et al.* 2018). A previous study in Nigeria (Ekpunobi *et al.* 2019) showed that some clays have chemical properties that allow their use for heavy metals removal. Additionally, it has been demonstrated that during the ceramic pot's firing process adsorbents like carbon are produced (Chaukura *et al.* 2020b). Therefore, CPFs have the potential to reduce the microbiological and chemical risk associated with water supply systems.

Although some types of arsenic are strongly adsorbed by naturally available clays, iron and manganese oxides/hydroxides and organic matter, the low arsenic removal reached in this study could also be attributed to the water's near-neutral pH, which reduces the adsorption of arsenic on metal oxides (Kumar *et al.* 2019).

Previous studies have shown that the removal of As(III) and As(IV) is the highest in pH values between 6.1 and 5.2 (Jadhav *et al.* 2015). The presence of arsenic in water supplies has been recognized worldwide as an important issue. Therefore, it is recommended to implement studies evaluating the different forms of arsenic removal in CPFs, taking into account the effects of pH.

Regarding the phenolic compound removal, Figure 4 shows the results during the study of the filtration systems by means of box and whisker plots. The average removal efficiencies were 78.7% for the CPF and 96.4% for the CPF + GAC-Z. During phase 1, the removal efficiencies were between 98 and 100% for the CPF filter and 100% for the CPF + GAC-Z filter. It is evident that in phase 2 there was a phenol removal efficiency reduction, mainly in the CPF system (64–94%), compared with the CPF + GAC-Z filter (87–99%). Besides, it was observed that in a higher 2,4,6,-trichlorophenol spiked water concentration the efficiency in the CPF filter increased, while in the CPF + GAC-Z the efficiency was more stable and higher. The high removal efficiency for CPF + GAC-Z shows the positive influence of the post-filtration column on the phenolic compound removal.

The average phenolic compound effluent concentration of the CPF filter was 0.4 mg/L, which is higher than the recommended limit value established by the WHO for 2,4,6-trichlorophenol (<0.2 mg/L). In contrast, the average effluent phenol concentration of the CPF + GAC-Z filter was 0.08 mg/L, which is below the recommended limit.

The ANOVA test confirmed that there were significant differences between the two systems for the phenolic compound removal (df = 3, F = 10.75, p < 0.001), and Tukey's contrast test showed the differences between CPF and CPF + GAC-Z

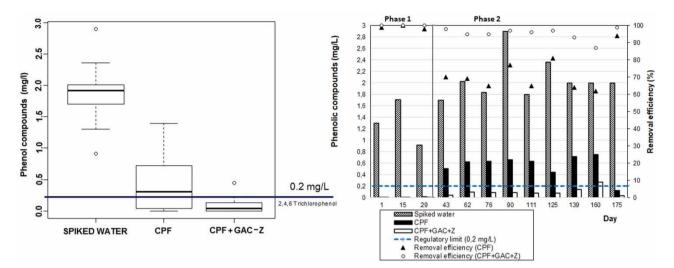


Figure 4 | Variation of the concentration and the removal efficiencies of phenolic compounds during the study. The horizontal line indicates the limit prescribed by WHO (2017).

(SE = 0.110331, p < 0.001). These results evidenced that the post-filtration column improved the removal of phenolic compounds.

This is in agreement with reports of an activated carbon efficiency of 80% for the removal of different organic constituents (WHO 2017), an efficiency that can be increased up to 98% when it is combined with iron oxide nanoparticles (Anku *et al.* 2017). Also, the use of zeolites has shown efficiency for the removal of chemical constituents due to its ion exchange capacity and adsorption properties (Akhigbe *et al.* 2016; Anku *et al.* 2017).

In general, the results are evidence of the improvement of CPFs when the post-filtration column with activated carbon and zeolite is implemented, such as in the CPF + GAC-Z system. However, further research is needed in order to evaluate arsenic removal at different pH values. This means that the choice of the CPF + GAC-Z mainly depends on the type of chemical risk posed by the water source, the economic resources of the user and the additional activities needed to maintain and clean the system.

It should be noted that the adsorption capacity of the systems could be attributed to the materials of the ceramic pot, as suggested by Chaukura *et al.* (2020b). To increase the efficiency of the systems, it is recommended to take this into account and evaluate the combination of clay with other adsorbent materials like activated carbon and nanoparticles (e.g. iron-based nanoparticles for arsenic removal, and carbon- and aluminum-based nanomaterials for fluoride removal), as recommended by Prathna *et al.* (2018). These materials could be evaluated using the same post-filtration column implemented in this study.

During the 175 days, the total volume of filtered water was 1,313 L. In this period, there was no evidence of the sorption capacity deterioration of the ceramic pot and the filtration column. However, it is well known that the adsorption process depends on the material's useful life. In particular, the life service of the GAC depends on the capacity of the carbon used and the contact time between the water and the carbon, as well as the water source characteristics and the chemical properties (water solubility and octanol–water partition coefficient) (WHO 2017).

Taking into account that in these filtration systems the effectiveness is eventually lost, depending upon the types of contaminating chemicals and their concentrations in the water (WHO 2017), it is proposed to implement a longer term study to evaluate the exhaustion of the ceramic pot and the filtration column. This is important because once the filtration system saturation point is reached, they can become a source of contaminants leaching out in the filtered water. Once this condition is reached, the options to renew the treatment system could be the regeneration of the filtration column and the replacement of the ceramic pot.

It is worth noting that the feasibility of CPF use for the production of water supply in geographically dispersed communities without safe water supply systems has been proved. Previous studies showed its effectiveness in ensuring the microbiological quality of water supplies (Brown & Sobsey 2010; Mwabi *et al.* 2011; Akosile *et al.* 2020). Respecting the use of the post-filtration column, the study carried out by Rivera-Sánchez *et al.* (2020) did not find statistically significant differences between the systems CPF and CPF + GAC-Z in terms of the microbiological removal efficiency, despite that the post-filtration column

media were impregnated with colloidal silver. This was attributed to the large pore size (>1 μ m) (WHO 2017). This allows us to conclude that in the CPF + GAC-Z, the main role of the ceramic pot is the microbiological risk reduction and that the post-filtration (GAC-Z) improves the chemical characteristics of the filtered water.

Based on the results obtained in this study and those reported in previous studies, it can be stated that the CPFs are also a viable option in cases where there are microbiological and chemical risks caused by anthropogenic activities. The use of a post-filtration column in the CPF + GAC-Z systems improves the efficiency of the traditional CPF, by increasing the removal of organic substances and lead. This means that the choice of the CPF + GAC-Z can depend on the type of chemical risk posed by the water source, the economic resources of the user and the additional activities needed to maintain and clean the system. It is recommended to carry out additional studies such as the EPA Challenge Test Water (turbidity at 30 NTU) to evaluate the post-filtration column performance in terms of the biofilm formation risk as well as operative aspects like cleaning and durability, as recommended by Pérez-Vidal *et al.* (2020). In addition to the above, additional studies can evaluate the influence of aspects such as the ceramic pot components and the burning temperature on the adsorption capacity of the ceramic pot.

CONCLUSIONS

- 1. The traditional CPF showed average removal efficiencies for Hg, Pb, As and phenolic compounds of 91.5, 92.1, 50.2 and 77.3%, respectively. Although the removal efficiencies in the CPF + GAC-Z (92.5% Hg, 98.1% Pb and 52.3% As) were higher for the removal of chemical constituents, there were no statistically significant differences between the two filter models. This is evidence of the importance of the ceramic pot for heavy metal removal. In contrast, the removal of phenolic compounds was higher (95.9%) in the CPF + GAC-Z filter, with statistically significant differences between the filter models. This is evidence of the contribution of the post-filtration column for the effective removal of the organic contaminants.
- 2. The results are evidence of the potential of CPF and CPF + GAC-Z systems to reduce the chemical risk associated with water supplies, mainly by Hg and Pb. However, the As concentration of the filtered water in both systems was higher than the regulatory limit, showing potential risk of As in the treated water by these systems.
- 3. The traditional CPF for water treatment supply is a viable option for the removal of microbial contaminants as has been demonstrated in previous studies. This study demonstrated that these systems can be used also for the removal of chemical contaminants. Additionally, the inclusion of a post-filtration column improves the performance of the systems reducing the potential chemical risk and thereby producing a safe water supply.

ACKNOWLEDGEMENTS

This research was funded by the Universidad Santiago de Cali and the company EKOFIL LTDA (manufacturer of the Ekoplast filters) (grant no. 711-621117-048). The authors thank Nienke Schellinkhout-Diaz and Gavin Howard for facilitating the writing of the manuscript. We also thank Mr Javier Moreno Gomez, founder of EKOKIL LTDA, for his support during the experimental phase.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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First received 9 February 2021; accepted in revised form 25 July 2021. Available online 9 August 2021