Science of the Total Environment 566–567 (2016) 368–377

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Multilevel modeling of retention and disinfection efficacy of silver nanoparticles on ceramic water filters

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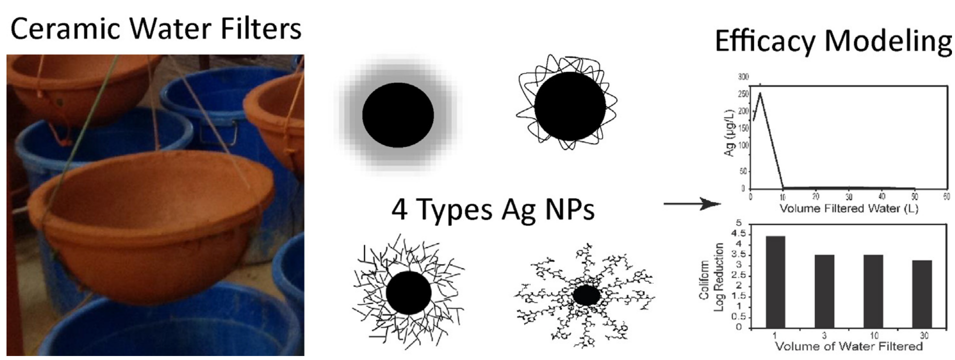
HIGHLIGHTS

- Multilevel modeling is useful for analysis of environmental data.
- Ag NP stabilizer does not significantly affect total Ag release from CWFs.
- Water type significantly affects the release of total Ag from CWFs.
- BPEI silver nanoparticles may provide improved coliform removal.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 2 March 2016
Received in revised form 11 May 2016
Accepted 12 May 2016
Available online xxxx

Editor: D. Barcelo

Keywords:
Silver nanoparticles
Multilevel model
Disinfection
Filtration
Ceramic water filters
Environmental engineering

ABSTRACT

This research examined how variations in synthesis methods of silver nanoparticles affect both the release of silver from ceramic water filters (CWFs) and disinfection efficacy. The silver nanoparticles used were stabilized by four different molecules: citrate, polyvinylpyrrolidone, branched polyethylenimine, and casein. A multilevel statistical model was built to quantify if there was a significant difference in: a) extent of silver lost, b) initial amount of silver lost, c) silver lost for water of different quality, and d) total coliform removal. Experiments were performed on location at Pure Home Water, a CWF factory in Tamale, Ghana using stored rainwater and dugout water (a local surface water). The results indicated that using dugout vs. rainwater significantly affects the initial (p-value 0.0015) and sustained (p-value 0.0124) loss of silver, but that silver type does not have a significant effect. On average, dugout water removed 37.5 μg/L more initial silver and had 1.1 μg/L more silver in the filtrate than rainwater. Initially, filters achieved 1.9 log reduction values (LRVs) on average, but among different silver and water types this varied by as much as 2.5 LRV units. Overall, bacterial removal effectiveness was more challenging to evaluate, but some data suggest that the branched polyethylenimine silver nanoparticles provided improved initial bacterial removal over filters which were not painted with silver nanoparticles (p-value 0.038).

Abbreviations: BPEI, branched polyethylenimine; CWF, ceramic water filter; DALYs, disability adjusted life years; LRV(s), log reduction value(s); Ag, silver; $AgNO_3$, silver nitrate; $NaBH_4$, sodium borohydride; TEM, transmission electron microscopy; PVP, polyvinylpyrrolidone; PPCC, probability plot correlation coefficient; PHW, Pure Home Water; WHO, World Health Organization.

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http://dx.doi.org/10.1016/j.scitotenv.2016.05.076

1. Introduction

The objective of this research is to understand how variations in synthesis methods of silver nanoparticles affect both the release of silver from ceramic water filters (CWFs) and disinfection efficacy. Silver nanoparticles are added to CWFs as a measure to protect human health and prevent biofilm growth from reducing the flowrate of the filter. The longevity of silver coatings on CWFs is not well understood. Failure to understand the fate of the silver puts users at risk of either decreased disinfection efficacy, due to total silver loss, or ingestion of silver concentrations above the World Health Organization's (WHO) acceptable limit of 0.1 mg/L, due to periodic large releases. This study links experimental data collected at a CWF factory with multilevel statistical modeling to quantify if there is a significant difference among four different types of silver nanoparticles in: a) extent of silver lost, b) initial amount of silver lost, c) silver lost for rainwater vs. dugout water (a local surface water source), and d) total coliform removal. Understanding how water type and silver type affect silver loss is a crucial first step to ensure that communities consistently obtain safe drinking water.

Large-scale efforts to provide global access to clean drinking water have been an international priority since the Millennium Development Goals were established in 2000 (Millennium Project, 2006). One household water quality intervention that has been particularly well received for its ease of use, cultural acceptance, and effectiveness is CWFs. After Hurricane Mitch hit Honduras in 1998, the organization Potters for Peace began widespread distribution of the CWF technology. Potters for Peace estimates that 46 independent organizations (in 18 different countries) produce variations of their CWF design (Potters for Peace, 2015).

Filters are made from local earthenware clays and a combustible material, such as sawdust or rice husks, which is burned out during the firing process, thereby leaving a porous structure. CWFs, which typically hold approximately 10 L of water in the pot, filter water at average rates of 1 to 5 L/h when the filter is clean and the feed water is not highly turbid (Rayner et al., 2013). Water is then stored in an encased plastic receptacle with a tap until used (Fig. 1).

Ren et al. (2013) found that CWFs are 3 to 6 times more cost-effective at reducing waterborne diarrheal illness than centralized water treatment systems in developing countries; their study used data for centralized water treatment systems in South Africa. Cost-effectiveness was evaluated via aversion to disability adjusted life years (DALYs) and water price. In the same study, they also found that CWFs positively impact human health because they contribute lower particulate matter emissions over the course of their lifecycle than centralized water treatment and distribution systems. In a separate study, using a meta-analysis focused on cost and aversion to DALYs, CWFs were also shown to yield a larger health impact (reduction of diarrhea) over lower cost household water treatment interventions such as chlorination and solar disinfection (Clasen and Haller, 2008).

Fig. 1. Typical ceramic water filter. Pure Home Water 2014 AfriClay filter and safe storage container. Note that the filter is suspended inside at the top of the storage container in the picture on the right.

Many CWF factories paint or dip the filters in silver nanoparticle solutions to provide disinfection. Numerous researchers have demonstrated that silver is effective at inactivating bacteria in solution (AshaRani et al., 2009; Auffan et al., 2009; Carlson et al., 2008; Eckhardt et al., 2013; El Badawy et al., 2011). Previous studies have also established that filters painted with silver nanoparticles provide increased bacterial removal versus bare filters (Bielefeldt et al., 2009; Oyanedel-Craver and Smith, 2008; PATH, 2012). However, some of these studies found a rapid loss of the silver, resulting in reduced disinfection efficacy after multiple batches of dirty water.

Research indicates that toxicity is related to the surface charge of the silver nanoparticle (El Badawy et al., 2011). None of the studies that focused on the silver applied to CWFs considered the synthesis method for the silver nanoparticles, even though several different techniques/molecules are used to control the shape, size, and surface charge of nanoparticles. It is unknown how different stabilizing molecules used in the production of silver nanoparticles affect attachment and detachment of the silver to the ceramic, and as a result the effectiveness and lifespan of the water filter. This study is the first of its kind to investigate the effect of molecular stabilizing agents on silver retention and disinfection efficacy on CWFs.

2. Materials and methods

The data used for this study were collected during field work at Pure Home Water (PHW), a filter factory located in Tamale, Ghana. Filtration experiments were performed on 15 filters divided into five different treatment scenarios (three filters each for four different types of silver nanoparticles and three filters without silver as a control for the bacterial portion of the work). To quantify filter effectiveness and lifespan, bacterial removal and total silver concentrations were monitored in the filtered water at 4 to 6 different time points (volumes of filtered water). Further, the experiments were performed using two different water sources: stored rainwater and water from a local surface water source called a dugout.

2.1. Nanoparticle synthesis & characterization

Four types of silver nanoparticles were used in this study. Polyvinylpyrrolidone (PVP), branched polyethylenimine (BPEI), and citrate stabilized silver nanoparticles were synthesized at The University of Texas at Austin (USA) and transported to PHW. The fourth type was purchased by PHW from Argenol Laboratories (Zaragoza, Spain). The colloidal silver from Argenol is routinely applied by PHW to the filters prior to distribution and it is stabilized by a casein molecule (Zhang and Oyanedel-Craver, 2011). Transmission electron microscope (TEM) images of representative PVP, BPEI, and citrate stabilized silver nanoparticles can be found elsewhere (Mikelonis et al., 2016), along with synthesis methods, cleaning procedures, stabilizer thickness measurements, and particle zeta potential values. As in Mikelonis et al. (2016), the citrate stabilized silver nanoparticles were synthesized according to Gorham et al. (2012), but with several modifications to the procedure. The prescribed synthesis method specifies bringing 400 mL of water to a rapid boil, adding 1.69 mL of a 58.8 mM $AgNO_3$ solution with 2.92 mL of a 34 mM sodium citrate solution, and dropwise addition of 2 mL of a 100 mM $NaBH_4$. Instead of these specific volumes, all quantities were multiplied by 10. The original method also prescribes stirring for 30 min at a slow boil, turning off the hot plate, and allowing to cool until room temperature, but for this work the solution was boiled rapidly over 10 h to concentrate the solution before cooling to room temperature. These modifications were made to facilitate transport to Ghana. The Argenol Ag nanoparticles (Fig. 2) were found to have an average diameter of 7 nm with a standard deviation of 4 nm, similar to those found earlier for the other Ag nanoparticles synthesized at UT Austin (average diameters for BPEI, Citrate, and PVP silver nanoparticles were estimated