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Research Paper

Lead-lag series and staged parallel operational strategies improve the performance and cost-effectiveness of bonechar for control of fluoride in groundwater

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ABSTRACT

Affordable, locally managed, decentralized treatment technologies are needed to protect health in resource-poor regions where communities consume groundwater containing elevated levels of fluoride (F). Bonechar is a promising low-cost sorbent for F that can be produced using local materials and simple pyrolysis technology. However, the sorption capacity of bonechar is low relative to the quantities of F that must be removed to meet health criteria (typically several mg/L), especially at pH typical of groundwaters containing high levels of geogenic F. This necessitates large bonechar contactors and/or frequent sorbent replacement, which could be prohibitively costly in materials and labor. One strategy for improving the feasibility of bonechar water treatment is to utilize lead-lag series or staged parallel configurations of two or more contactors. This study used column testing to quantify potential benefits to bonechar use rate, replacement frequency, and long-run average F concentration in treated water of lead-lag series and staged parallel operational modes compared with single contactor mode. Lead-lag series operation exhibited the largest reduction in bonechar use rate (46% reduction over single contactor mode compared with 29% reduction for staged parallel) and lowest long-run average F levels when treating central Mexican groundwater at pH 8.2 containing 8.5 mg/L F.

Key words | bonechar, fluoride, groundwater, household water treatment, Mexico, sorption

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INTRODUCTION

The negative health impacts associated with consuming elevated levels of fluoride (F) include painful and debilitating skeletal and dental fluorosis (WHO 2011). It is estimated that over 200 million people worldwide are exposed to harmful levels of F by drinking groundwater that exceeds the World Health Organization guideline value of 1.5 mg/L (Amini *et al.* 2008; WHO 2011). Many of these people live in low resource settings including dispersed rural settlements lacking infrastructure. One such region is Guanajuato state, central Mexico, where F levels in community water sources can exceed 20 mg/L, although values ranging from 2 to 8 mg/L are more common (Caminos de Agua 2018). Affordable, locally managed decentralized water treatment technologies are needed to protect public health in this region.

Pyrolyzed animal and fish bone material (bonechar) has been investigated by many researchers as a low-cost sorbent for uptake of F from groundwater. It can be produced using materials that are commonly available in rural areas, as has been demonstrated in Ethiopia, Kenya, and Tanzania (Mjengera & Mkongo 2002; Abaire *et al.* 2009; Brunson & Sabatini 2009). However, the F sorption capacity of unmodified bonechar is relatively low – typically 1–2 mg/g at equilibrium liquid concentrations ~1.5 mg/L) (Brunson & Sabatini 2009; Yami *et al.* 2015, 2017) – especially at elevated pHs (8–9) typical of groundwaters containing geogenic F (Medellin-Castillo *et al.*

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2007). Various methods have been pursued for increasing the F sorption capacity of bonechar, such as thermal and chemical activation using strong acids and bases, and impregnation with aluminum salts or oxides (Brunson & Sabatini 2014; Yami et al. 2016; Nigri et al. 2017). The drawbacks of these bonechar production process modifications include the additional challenges, costs, and hazards associated with acquiring and safely handling reagents and treating the waste stream under rural and developing community circumstances.

A different strategy for improving F control using bonechar that is implementable at the local level in developing communities is to modify the operational regime of fixed-bed contactors. Lead-lag operation of two fixed-bed contactors in series as well as staged parallel operation of multiple contactors with blended effluent have been shown to improve (i.e., reduce) sorbent use rates for control of trace pollutants in water treatment (Dvorak et al. 2008; Denning & Dvorak 2009; Corwin & Summers 2012; Stewart et al. 2013). In lead-lag configuration, two contactors are operated in series until the concentration of the pollutant of interest in the effluent of the second (lag) column rises to the treatment objective (e.g., 1.5 mg/L F). The sorbent is then replaced in the first (lead) contactor and the order of the contactors reversed such that the former lead contactor becomes the lag contactor. This allows for utilization of some of the remaining sorption capacity in the former lag column. This capacity goes unused under conventional single-contactor operational regimes since the sorbent must be replaced as soon as the treatment objective is reached.

In staged-parallel operation, the media is replaced in two or more contactors in a staggered periodic manner such that one or more contactors are producing water below the treatment objective at any given time. The effluents from individual contactors are blended so that the system effluent remains below the treatment objective. Under staged parallel management, one or more contactors are allowed to operate beyond the treatment objective in their individual effluent(s), utilizing sorption capacity that would go unused under single-contactor operational regimes.

Most published studies of F uptake by bonechars have been conducted in batch-test format using powdered sorbent. A comparatively small number of studies have quantified F uptake from real or simulated groundwater by raw or modified bonechars in fixed-bed (column) format using granular sorbent (Brunson & Sabatini 2014; Nigri et al. 2017; Yami et al. 2017). To our knowledge, no studies have been published investigating different operational modes for fixed-bed contactors using bonechar sorbent. Therefore, the objective of this study was to quantify the extent to which lead-lag series and staged parallel operational strategies could improve the performance and cost-effectiveness of bonechar for controlling F in groundwater over conventional single-contactor operation.

METHODS

Bonechar preparation and characterization

Cow bones obtained from a slaughterhouse located near San Miguel de Allende, Guanajuato, Mexico were cut into pieces and dried in the sun for several days. Dried bone pieces were packed into a 95 L (25 gal) lidded steel drum retort and heated to approximately 570 °C over a 2-hour period (heating rate ~4.5 C/min). The retort was allowed to remain at ~570 °C for an additional 45 minutes and then cooled rapidly with water to handling temperature. More details of bonechar production are provided in Figures SI-1 and SI-2 in Supplementary information (available with the online version of this paper). Bonechar pieces were ground by hand with a mortar and pestle and wet-sieved to collect the fraction retained between #8 and #30 US Standard sieves (2.36 and 0.60 mm, average particle diameter 1.29 mm) for use in column experiments.

Images were collected of bonechar granules using a Hitachi S3200N variable pressure scanning electron microscope (VP-SEM) located at the NC State University Analytical Instrumental Facility. Relative elemental abundances on the surfaces of bonechar granules were determined using energy dispersive X-ray spectroscopy (EDS). Bonechar BET (Brunauer–Emmett–Teller) surface area was determined by N_2 adsorption using a Quantachrome Autosorb-1 MP.

Column experiments

Columns were constructed from commercially available cartridge filter housings (purchased from a local hardware store in San Miguel de Allende, grupodonpedro.com) with refillable inserts (AMI Filters, part number C-C2510-EP, appliedmembranes.com). Photographs and diagrams of the column hardware are shown in Figure SI-3 in the Supplementary information (available online). Internal dimensions of the inserts were 7.01 cm diameter by 18.0 cm depth with an empty bed volume of 695 mL. Inserts were filled with 459 g of granular bonechar for a bed density of 660 g/L. Columns were connected in series using PTFE tubing and fittings. Ports were located between columns 1 and 2 and after column 2 for collecting column effluent samples. Replicate experimental setups ('A' and 'B') consisting of two columns in series, each, were fed from the same influent storage tank by gravity. Ball valves installed after the second column of each setup were used to control the flow rate at ~18 mL/min. This flow rate was chosen to produce ~25 L per day, e.g., to provide one household of five members 5 L of treated water per person per day. The flow rate of setup A was 18.2 mL/min (standard deviation 2.1 mL/min) for an average empty bed contact time (EBCT) of 38.2 minutes for the individual columns and an EBCT of 76.3 minutes for both columns in series. The flow rate of setup B was 18.1 mL/min (standard deviation 1.8 mL/min) for an average EBCT of 38.4 minutes for individual columns and an EBCT of 76.8 minutes for both columns in series. The test water (8.5 mg/L fluoride, pH 8.2) was collected from a community well in the village of Ex Hacienda de Jesus in Guanajuato state, Mexico. For experimental setup A, samples were collected for fluoride quantitation until the effluent of column 2 reached ~2.5 mg/L (20 days). For setup B, the experiment was discontinued when the effluent of column 2 reached ~1 mg/L (18.5 days) due to sediment fouling of the column and inability to maintain flow rate at 18 mL/min. Fluoride in influent and effluent water samples was quantified using a portable colorimeter (Hach DR850) and SPADNS 2 reagent method.

RESULTS AND DISCUSSION

Bonechar characterization

The N_2 BET surface area of the bonechar used in this study was 114 m^2/g. This value is similar to bonechars used in other studies – for example, 111 m^2/g (Brunson & Sabatini 2009), 100 m^2/g (Cheung *et al.* 2005), and 104 m^2/g (Medellin-Castillo *et al.* 2007). EDS spectra of different regions of bonechar surfaces indicated relative elemental abundances consistent with a mixture of bone mineral (hydroxyapatite, $Ca_5(PO_4)_3(OH)$) and carbonized organic matter (Figure 1). Pure hydroxyapatite is 39.9% Ca, 41.4% O, 18.5% P, and 0.2% H by weight. The bonechar regions analyzed by EDS ranged from 14.2 to 23.4 weight-% C. Additional SEM images of bonechar granules are provided in Figure SI-4 in the Supplementary information (available online).

Column experiments

Figure 2(a) displays fluoride breakthrough curves for columns 1 (open symbols) and 2 (closed symbols) from the experimental setup A. Similar data were obtained using setup B and are provided in Figure SI-5 in the Supplementary information (available online). The experimental setup A column test was carried out for just over 20 days, at which time the capacity for F uptake by the bonechar in column 1 was exhausted and effluent from column 2 had reached ~2.5 mg/L F.

Lead-lag series operation

A lead-lag operational strategy employing contactors in series can improve sorbent utilization compared with single contactor performance. Under lead-lag mode, two contactors in series are operated until the treatment objective is reached in the effluent of the second (lag) contactor. At that time, the sorbent medium is replaced in the first (lead) contactor and the order of the contactors reversed such that the former lead contactor becomes the lag contactor and vice versa.

As described by Corwin & Summers (2012), the sorbent use rate (or cycle time) can be estimated for two contactors operated in lead-lag mode using breakthrough curve data from a single experiment employing two columns with the same EBCT in series as shown in Figure 2(a). Use rate is determined by integrating the sorbed mass of contaminant on the lead column (shaded *Area* A_{LL} in Figure 2(a)) plus the mass in the lag column effluent to the point at which the treatment objective is reached (shaded *Area B* in Figure 2(b)). The unshaded area in Figure 2(a) represents the mass sorbed on

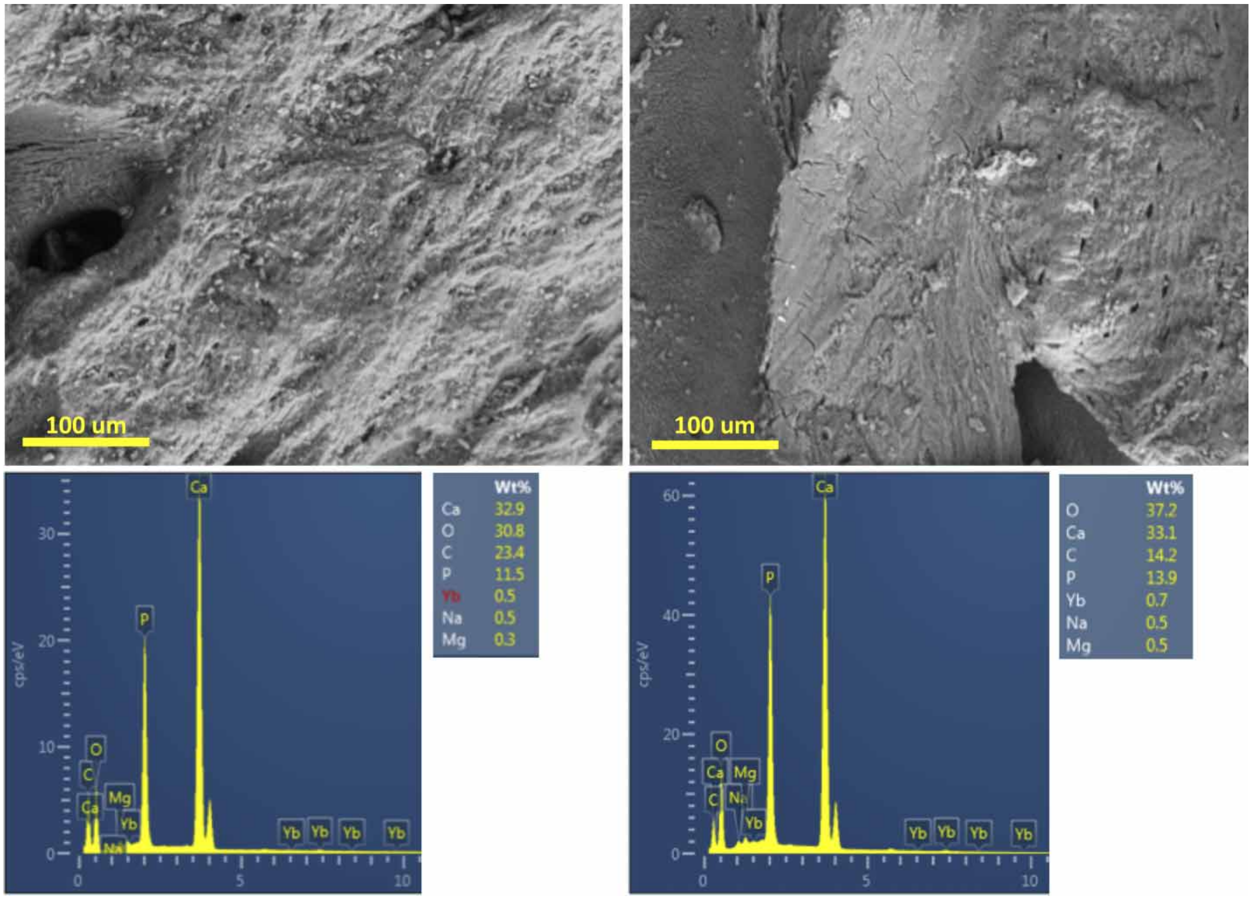

Figure 1 | VP-SEM images and EDS spectra of bonechar granules.

the lag column where F will continue to be sorbed during the next cycle when it becomes the lead column.

Sorbent use rate under lead-lag series operation (SUR_{LL}) in units of g/L is estimated using Equation (1):

$$SUR_{LL}\left(\frac{g_{sorbent}}{L}\right) = \frac{C_0\left(\frac{mg}{L}\right) \times EBCT(\min) \times bed\ density\left(\frac{g}{L}\right)}{1440\left(\frac{min}{d}\right) \times \left[Area\ A_{LL}\left(\frac{mg}{L} \times day\right) + Area\ B\left(\frac{mg}{L} \times day\right)\right]} \quad (1)$$

Here, C_0 is the influent concentration of the contaminant of interest in mg/L, *EBCT* is the EBCT of a single contactor in minutes, and the bed density of sorbent is inputted in g/L. The sorbent use rate for a single contactor (SUR_{SC}) is calculated similarly (Equation (2)) by substituting the integrated sorbed mass of contaminant up to the point that column 1 reaches the treatment objective (*Area* A_{SC}, for 'single contactor' in Figure 2(a)) and omitting *Area B*.

$$SUR_{SC}\left(\frac{g_{sorbent}}{L}\right) = \frac{C_0\left(\frac{mg}{L}\right) \times EBCT(\min) \times bed\ density\left(\frac{g}{L}\right)}{1440\left(\frac{min}{d}\right) \times \left[Area\ A_{SC}\left(\frac{mg}{L} \times day\right)\right]} \quad (2)$$

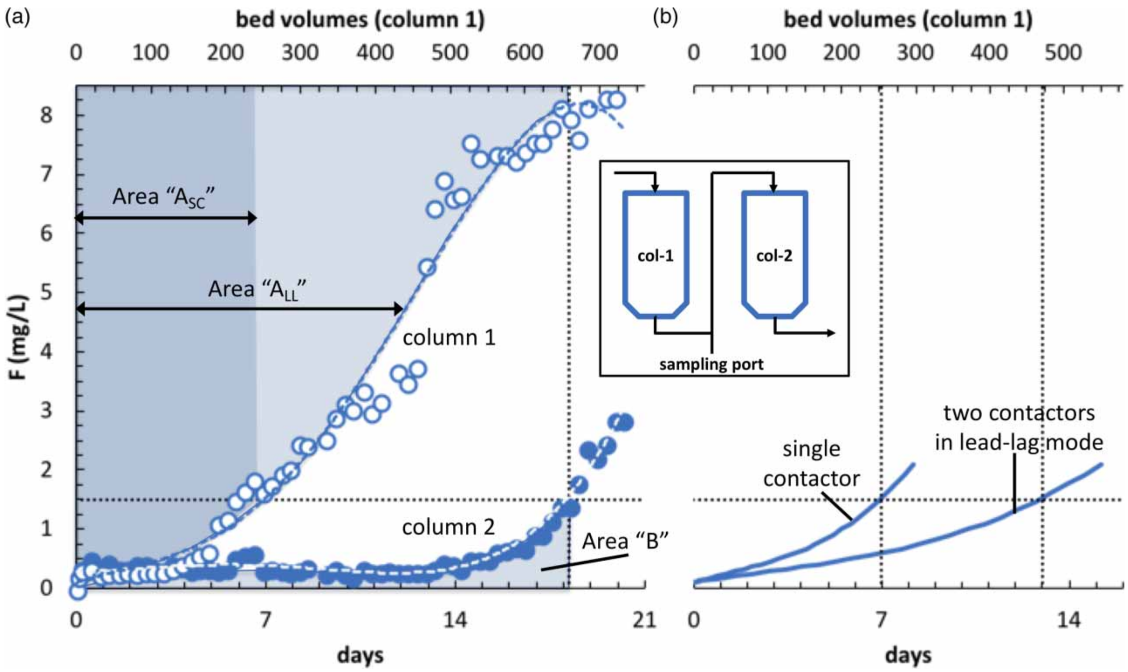

Figure 2 | Fluoride breakthrough curves from bonechar columns in series (experimental setup A) with column throughput shown in units of time (days, lower x-axis) and bed volumes (upper x-axis) (a). The dashed lines indicate fourth-order polynomial fits to the data. The vertical dotted line indicates column throughput in days and bed volumes when the F concentration in the effluent from column 2 rose to the 1.5 mg/L treatment objective. (b) The predicted cycle time is indicated in days and bed volumes for a single contactor and for two contactors operated in lead-lag series mode (discussed in the next section). The inset shows the treatment flow diagram.

Using the F breakthrough data in Figure 2(a), the SUR_{SC} with an *EBCT* of 38.2 minutes was 2.8 g of bonechar per liter of water treated. For two contactors operated in lead-lag series mode, the sorbent use rate (SUR_{LL}) was estimated to drop to 1.5 g/L – a reduction of approximately 46%. Another way of expressing this is to compare the estimated cycle time (replacement frequency) of two lead-lag series contactors with that of a single contactor. This is illustrated in Figure 2(b), where the cycle time is predicted to increase from approximately 7 days for a single contactor to approximately 13 days for two contactors operated in lead-lag series mode. This is equivalent to extending the bed life of a single contactor from approximately 250 bed volumes to approximately 475 bed volumes (Figure 2(b)).

For experimental setup B, SUR_{LL} was also estimated to be 1.5 g/L (Figure SI-5 in the Supplementary information). SUR_{SC} was 3.7 g/L and cycle times for single contactor and lead-lag series contactors was estimated to be 5.5 and 13.5 days, respectively.

Staged parallel operation

Operating contactors in staged parallel configuration can also lower sorbent use rates (Denning & Dvorak 2009; Corwin & Summers 2012; Stewart *et al.* 2013). At startup, two (or more) contactors with fresh media are operated in parallel and their effluents blended. When the blended effluent concentration of the pollutant of interest reaches the treatment objective, the sorbent in one contactor is replaced. The other contactor is allowed to continue operating beyond the treatment objective in its individual effluent. When the blended effluent again rises to the treatment objective, the medium in the contactor that has been in service the longest is replaced and the cycle resumed. After several cycles the replacement frequency converges on a long-run value.

Figure 3 displays a simulated operation of two contactors in staged parallel mode. The polynomial fit to column 1 F breakthrough data shown in Figure 2(a) was used to simulate several cycles of operation. After a few simulation

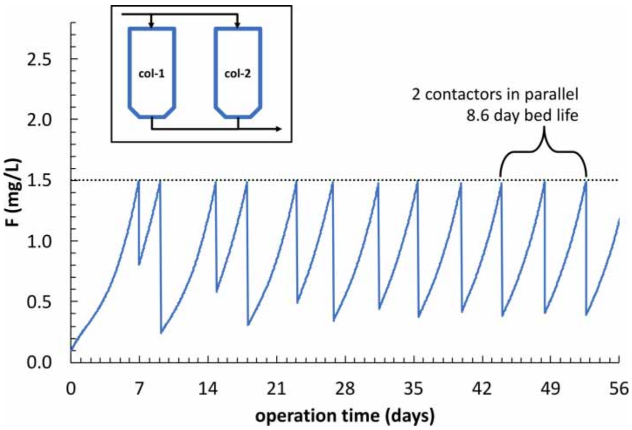

Figure 3 | Simulated operation of two bonechar contactors (*EBCT* 38.2 minutes, F C_0 = 8.5 mg/L) in staged parallel mode. The polynomial fit to column 1 F breakthrough data (Figure 2(a)) was used to simulate several cycles of operation using MS Excel. The inset shows the treatment flow diagram.

cycles the bed life converged on 8.6 days, corresponding to a sorbent use rate of 2.0 g/L.

Comparison of operational strategies

Table 1 summarizes parameters of interest for selecting bonechar contactor operational strategies for fluoride control in groundwater.

In this study, simulations developed from bonechar column data indicated that lead-lag series operation is expected to provide improved sorbent utilization (i.e., lower use rate and less frequent media replacement) compared with single contactor and staged parallel operational strategies under the conditions studied. This result is not surprising considering the onset and shape of the column 1 breakthrough curves shown in Figure 2(a) (and Figure SI-5)

Table 1 | Summary of parameters of concern for single contactor, lead-lag series contactors, and two-staged parallel contactors operational strategies for fluoride control (C_O = 8.5 mg/L) by bonechar sorbent

	Single contactor	Lead-lag series contactors	Two-staged parallel contactors
Sorbent use rate (g/L)	2.8 (3.7)	1.5 (1.5)	2.0
Replacement frequency (d)	7.0 (5.5)	13.0 (13.5)	8.6
Average effluent F concentration (mg/L)	0.70 (0.79)	0.39 (0.48)	0.88

Values estimated from data collected using experimental setup B are given in parentheses.

and the ratio of F treatment objective to influent F concentration. To explain further: the mass transfer zone (*MTZ*) is the portion of the column where sorption is taking place at a given point in time. Behind the *MTZ*, the liquid concentration of the sorbate of interest is equal to the influent concentration (i.e., $C/C_0 = 1$) and no further sorbate uptake is occurring. Ahead of the *MTZ*, the liquid concentration of the sorbate is 0 and the sorbent has not yet been exposed to the sorbate. Following previous research (Crittenden *et al.* 1987; Hand *et al.* 1989), a proxy for the breadth of the *MTZ* is the time elapsed between 5% and 95% breakthrough (i.e., $C/C_0 = 0.05$ and 0.95). The Lag period (not to be confused with lead-lag operation) was defined by Stewart and coworkers (Stewart *et al.* 2013) as the time elapsed until breakthrough reaches 5% ($C/C_0 = 0.05$). The ratio of *MTZ* to *Lag* (*MTZ:Lag*) reflects over what portion of the length of the column sorption is actively taking place. This ratio is a function of column length, *EBCT*, sorption kinetics, and background water characteristics. Increasing column length and/or *EBCT* and faster sorption kinetics correspond to decreasing *MTZ:Lag* ratios. The polynomial fit to column 1 data (Figure 2(a)) was used to estimate a *MTZ:Lag* ratio of 5.6 for this study. This is a relatively high value (Stewart *et al.* 2013), indicating that a significant portion of the bonechar column was occupied by the *MTZ*. When *MTZ:Lag* ratio is high, greater benefit to sorbent use rate from lead-lag series or staged parallel operation compared with single contactor operation is expected (Denning & Dvorak 2009; Stewart *et al.* 2013).

The ratio of target sorbate treatment objective to influent concentration (C_{TO}/C_0) influences whether lead-lag or staged parallel confers greater benefits to sorbent use rate (Dvorak *et al.* 2008; Denning & Dvorak 2009; Stewart *et al.* 2013). As C_{TO}/C_0 value decreases (i.e., a more stringent treatment objective is set), lead-lag operation is favored. In this study, $C_{TO}/C_0 = 0.18$. Thus, water sources with F concentrations >8.5 mg/L ($C_{TO}/C_0 < 0.18$) would be expected to gain additional *SUR* benefit from lead-lag operation. Conversely, water sources with F concentrations that slightly to moderately exceed 1.5 mg/L might derive greater *SUR* benefit from staged parallel column operation. Increasing the number of contactors in parallel would improve *SUR*. However, this adds additional up-front cost and system complexity. Also, the long-run average effluent concentration of F approaches the 1.5 mg/L WHO guideline value as the number of contactors is increased.

Average effluent F concentrations were estimated for single contactor and lead-lag series operation by integrating the areas under the column 1 and column 2 curves in Figure 2(a) and dividing by the volume throughput to treatment objective (Table 1). Average effluent fluoride concentration was estimated for staged parallel operation using simulation values in Figure 3 after the bed lifecycle had stabilized (after ~28 days). In this study, lead-lag series operation was predicted to provide the lowest time-averaged levels of F in drinking water (0.39 mg/L) compared with staged parallel and single contactor operation (0.88 and 0.70 mg/L, respectively) (Table 1). Comparing lead-lag with single contactor, the benefit in F reduction is conferred by the extended *Lag* period that allows the relatively long *MTZ* to pass through two sorbent columns in series versus a single column.

Recommendations for WaSH practitioners

In this study, significant effort was made to conduct column experiments under realistic field conditions by using locally available hardware for constructing bonechar contactors, bonechar generated in the field at the application scale, and groundwater from an affected rural Mexican community. Previous research demonstrated that conducting column studies using laboratory DI water or synthetic groundwater overpredicts F uptake by bone-derived adsorbents compared with real groundwaters evaluated under field conditions (Brunson & Sabatini 2014; Yami *et al.* 2017). Groundwater F concentrations as well as the presence and concentration of other dissolved species (e.g., anions such as phosphate and silicate; dissolved organic matter) that can compete with F for adsorption sites and/or block access to bonechar pores vary geographically and through time. Therefore, bonechar replacement frequencies reported here should not be universally applied to all household and community water treatment circumstances. Rather, this study provides a theoretical justification and experimental methodology for conducting rapid quantitative assessments of bonechars and groundwaters under representative field conditions to derive optimal site-specific operational strategies. The refillable contactors (~700 mL bed volume) and cartridge housings used in this study are of standard design and are relatively inexpensive and widely available. This system can be operated with a ~40 minute *EBCT* (per column) to provide ~25 L of treated water per day – potentially sufficient for one household. If the bonechar particle size and *EBCT* are held constant, this system can be scaled up to provide larger quantities of treated water (e.g., for a community) with similar expected F control and bed life for a given bonechar and groundwater combination (Crittenden *et al.* 2012).

CONCLUSIONS

This study used column tests to investigate operational strategies – lead-lag series and staged parallel modes – for improving the use rate of bonechar sorbent for control of excessive fluoride in drinking water. For well water from a central Mexican village at pH 8.2 containing 8.5 mg/L F, lead-lag series operation was predicted to decrease sorbent use rate, extend bed life, and provide lower average levels of F in drinking water compared with staged parallel and single contactor operational modes. The disadvantages of lead-lag operation compared to single contactor mode include greater startup cost (additional column hardware, plumbing, and media), and more complex management (swapping the order of contactors with each media replacement cycle). Staged parallel operation might result in lower sorbent use rates than lead-lag mode if F concentration in source water is close to the 1.5 mg/L treatment objective; however, this would need to be determined experimentally. In addition to enhancing sorption capacity by modifying bonechar production, management strategies such as lead-lag series operation of contactors can enhance the utility of bonechar sorbent for fluoride control in drinking water in low resource settings.

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