Assessment of biologically active GAC and complementary technologies for gray water treatment
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ABSTRACT
The reuse of gray water for applications ranging from irrigation to showering is a viable means to reduce net water demand when water supplies are stressed. The objective of this study was to investigate the treatment of gray water using biologically active granular-activated carbon (GAC) and complementary technologies. Technologies were challenged individually or in combination using a synthetic gray water formulation based on NSF/ANSI Standard 350. Specific technologies included: GAC; biologically active GAC (BAC); a newly developed intermittently operated BAC (IOBAC) process; ion exchange (IX); coagulation with a cationic polymer; microfiltration; ultrafiltration (UF); and multi-barrier combinations thereof. For control of organic contaminants such as surfactants, BAC and IOBAC performed well over test periods as long as 6 months. Combinations of IOBAC treatment with coagulation pretreatment and UF post-treatment resulted in sustained chemical oxidant demand and turbidity value reductions in excess of 90 and 99.5%, respectively. Such an approach would be useful for gray water treatment for low tier applications like irrigation or toilet flushing, or as a pretreatment system upstream of reverse osmosis (RO) membranes and/or advanced oxidation processes for high tier reuse applications such as showering.

Key words | biofiltration, gray water, water reclamation, water reuse

INTRODUCTION
As water stress continues to increase throughout the world, the capability to treat and reuse gray water for a broad range of reuse applications will be needed. In the USA, reuse of gray water is generally limited to toilet flushing as the highest tier with respect to human exposure (USEPA 2012). In many states, guidance for gray water reuse has yet to be adopted (Sharvelle et al. 2013). However, in countries such as Australia, and in deployed scenarios such as contingency bases for the US military, direct reuse of gray water for higher tier applications, such as showering and/or laundring, is already being practiced (Fifty 2008). As a growing capability area, gray water treatment for a broad range of reuse activities will continue to be improved in terms of robustness and energy efficiency while still providing high quality water with low health risks. Because gray water has distinct features, further study of its treatment and customization of associated treatment technologies is necessary.

Gray water is defined herein as the waste water generated by bathroom sinks, showers, and laundry facilities (Christova-Boal et al. 1996; Little 2002; Al-Jayyousi 2003; Wilderer 2004). By this definition, it does not include waste water from kitchens, which has relatively high organic load and lower biological stability. Even when excluding...
kitchen wastewater, gray water represents a large fraction of the wastewater generated in residential, commercial, and military settings. In some of these cases, gray water can represent up to 70% of the wastewater generated (Fifty 2008). Recycling or reusing gray water could therefore drastically reduce requirements for potable water supply for a given location.

Previous studies on gray water composition and quality have shown that there is considerable variability in water quality with respect to source type (Holden & Ward 1999; Al-Jayyousi 2005; Jefferson et al. 2004; Winward et al. 2008), even when kitchen wastewater is not included in the comparisons. In general, gray water mixtures comprised of effluents from sinks, showers, and laundry typically exhibit organic and particulate loadings similar to those of municipal wastewaters. However, they differ from wastewater in nutrient content, classes of molecules, pathogen concentrations, and biodegradability. Consistent with the target ranges in NSF/ANSI Standard 350 for challenge testing of gray water treatment systems (NSF International 2011), influent gray water is typically characterized as having chemical oxidant demand (COD) from 200 to 400 mg/L, biochemical oxygen demand (BOD) from 100 to 200 mg/L, total suspended solids (TSS) from 80 to 120 mg/L, and turbidity levels from 50 to 80 nephelometric turbidity units (NTU). Thus, substantial water quality improvements are required to enable safe reuse for most activities. In addition to water quality challenges, gray water treatment systems need to be tolerant of variable, intermittent flows and loadings of organics and particulates. Further, gray water systems are often implemented in a distributed fashion, presenting additional constraints for automation, maintenance, physical footprint, and energy-efﬁciency due to less operator support and space at the household or building level.

The objective of the present study was to evaluate several candidate low-logistics water treatment technologies in terms of their ability to efﬁciently control particulate and organic contaminants in gray water. Speciﬁcally, the use of biologically active granular-activated carbon (GAC), biologically active granular-activated carbon (BAC), intermittently operated BAC (IOBAC), ion exchange (IX), coagulation with a cationic organic polymer, microﬁltration (MF), and ultraﬁltration (UF).

**METHODS**

A synthetic gray water solution was prepared for the experiments based on guidance in NSF/ANSI Standard 350 (NSF International 2011). The mixing ratio of bathing to laundry water formulations was used 2:1. Specific components and quantities are provided in Table 1. Water quality in the feed tank was monitored over the extent of the residence time in several experiments to conﬁrm consistent quality of the influent. A paddle mixer (1,500 rpm) was operated for 15 of every 90 minutes to minimize settling in the influent supply tank.

All water quality samples were either analyzed within 1 hour of sampling or were stored at 4°C until analysis within 12 hours. COD was measured according to Standard Method 5220 D, and 5-day BOD (BOD-5) was measured according to Standard Method 5210 B (American Public Health Association/American Water Works Association/
Turbidity was measured using an HF Scientific DRT-15CE turbidimeter that was calibrated against manufacturer-provided reference standards on each day of analysis. TSS was measured according to Standard Method 2540 D. Analyses for ammonia, nitrite, nitrate, and total Kjeldal nitrogen (TKN) were also performed periodically on the influent to ensure consistency with NSF/ANSI Standard 350 target ranges (NSF International 2011). Phosphorous concentration was measured using the Standard Method 4500-P B. Relative surfactant concentration was measured indirectly using a modified cylinder shake method (Stiepel 1944) that quantified the amount of foam after a fixed time of controlled sample agitation in a 25 ml sample. The associated calibration curve generated from six dilutions of gray water influent was reproducible and linear \( (R^2 = 0.992, n = 3) \). Relative surfactant concentrations measured using this method were consistent with those measured using Standard Method 5540C. Silt density index and low-pressure membrane fouling rate analyses were conducted at 30 psi using MF membranes and a test apparatus procured from Applied Membranes Incorporated (Vista, CA, USA).

For all GAC filtration studies, research-grade GAC media was obtained from API Inc. (Chalfant, PA, USA) and washed vigorously in deionized water prior to use. Additional assessments of the GAC were performed after cleaning the carbon. Specific surface area was measured using a NOVA 3200 BET analyzer (Quantachrome Corp, Boynton Beach, FL, USA). Additional GAC characterization analyses included solid-state nuclear magnetic resonance (NMR), total elemental analysis by energy-dispersive X-ray spectroscopy (EDX), dynamic light scattering, and potentiometric titration to determine zero point of charge. The activated carbon tested had a mean surface area of 532 m²/g. \(^{13}\)C-NMR studies confirmed a highly aromatic structure with no detectable signatures of alkyl, o-alkyl, or carboxylic functional groups, consistent with pyrolysis at high temperature. EDX analysis calculated the elemental distribution to be 85% C, 8.2% Si, 2.5% S, 1.5% Ti, 1.2% Ca, and less than 1% of other elements. Titration studies confirmed a single pKa at 7.5, with limited buffering capacity.

Baseline non-biologically active GAC breakthrough experiments were performed using vertically oriented 5-cm diameter PVC columns with a GAC-bed depth of 25 cm. Columns were operated in an upflow manner over a range of flow rates. Sample ports at 0, 5, 10, 15, and 20 cm were utilized to sample water quality as a function of depth in the column over time. Fresh GAC was used for each experimental condition. All experiments were performed at room temperature (21–23°C).

BAC filtration studies were performed using a 5-cm diameter PVC column with a bed depth of 20 cm. GAC media was prepared as described above, with the exception that the filter was pretreated with one bed volume (BV) of water containing bioseed culture. The bioseed culture was prepared by inoculating 250 ml of custom media with 0.1 g of commercial top soil (Scotts Premium) and 1 ml of secondary wastewater influent from the local wastewater treatment plant (Urbana, IL, USA). The custom media contained 0.5 g/L yeast extract and 0.5 g/L nutrient broth (both Difco), 5 ml/L of hand soap containing tricosan, and 5 ml/L of shampoo/conditioner. The seed culture was grown at room temperature for 20 hours prior to centrifugation at 2,000 rpm for 10 minutes and resuspension in 1 BV of deionized water. Seed culture suspension was applied to the GAC column at a flow rate of 1 BV per hour within 12 hours of starting the experiment. For the experiments, gray water was delivered to the BAC column at a flux value of 1.3 Lpm/m². Continuous upflow testing was performed for a period of 5 weeks after seeding the filter to evaluate sustained performance of the BAC system. Filter media was suspended and flushed once every 2 days using aeration under upflow conditions to dislodge biomass and debris in the filter.

As an alternative to conventional continuous BAC studies, a new IOBAC approach was investigated. The IOBAC filtration studies were performed as described for the BAC filtration studies, with the exception that the filters were challenged with a higher flux (4 Lpm/m²) for two abbreviated intervals each day, followed by periods of non-operation in which the filters were completely drained. IOBAC filter treatment volumes were equivalent to those used in the BAC studies but were loaded cyclically at higher rates for 4 hours, followed by an 8 hours bioregeneration period, such that the same daily loadings were achieved. Under this operating condition, the IOBAC filters were tested for over 120 days to assess the self-cleaning
bioregeneration effect for sustained filter capacity. Additional experiments with a taller (4×) column and equivalent volumetric loading rate were also performed.

IX experiments were performed in batch and column configurations. Tanex resin (Purolite), a mixed anion exchange resin that targets dissolved organic molecules with functionality similar to tannins, was the only resin tested in this study. Column experiments were performed at a loading rate of 5 BV/h. Due to incompatibility of Tanex with high concentrations of colloidal material, gray water for these experiments was pretreated with UF (0.05 μm pore size) to remove colloids, which reduced the influent COD by 40–50% and altered the gray water composition. Columns were regenerated with 10% sodium chloride brine solution after each test run.

Coagulation experiments were performed using a cationic polymeric coagulant, poly(acrylamide-co-diallyldimethyl-ammonium chloride) (PDM), obtained from Sigma-Aldrich. Jar tests were conducted in 150-ml cylindrical batch reactors using identical mixing conditions for rapid mix (650 rpm, 2 minutes), flocculation (150 rpm, 15 minutes), and settling (0 rpm, 120 minutes). In order to remove the resultant low-density flocs from solution, samples were centrifuged at 1,500 rpm for 10 minutes. Positive controls with no coagulant were subjected to the same process to account for any particulate or organics removal associated with the centrifugation step.

Membrane filtration studies were performed using hollow fiber polyether sulfone MF and UF filters from Sterlitech (Kent, WA, USA) with 0.2 μm and 0.05 μm absolute pore size, respectively. Prior to testing, membranes were soaked overnight in 10% methanol and washed several times. At least three cycles of filtration, backflushing, and chemical cleaning were performed for each test condition. Clean water flux was determined after running deionized water through the pre-wetted membrane for 15 minutes, alternating between forward flow and backward flow every 5 minutes. Membranes were then challenged with gray water (10 L) at 50 psi, and flux was monitored by quantifying the mass of effluent collected during 30 seconds time intervals for up to 30 minutes prior to backflushing. Each backflush consisted of running deionized water backward through the membrane for 5 minutes in order to remove loosely bound foulants from the membrane. After testing, a cleaning procedure included backflushing at 20 psi and rinsing with chlorinated water (200 ppm as Cl2).

RESULTS AND DISCUSSION

Influent water quality data for the gray water tests in this study are shown in Table 2. Average values, number of replicates (n), and 95% confidence intervals are presented. Also listed are relevant water quality parameter ranges provided in the NSF/ANSI Standard 350 guidance (NSF International 2011). In general, the gray water influent used in this study was consistent with the NSF 350 ranges for each parameter, with organic contaminant indicators BOD-5 and COD being on the higher end.

Prior to conducting studies of BAC, baseline studies were performed using non-biologically active GAC columns to assess gray water treatment performance in the absence of biodegradation (i.e., adsorptive removal of clean GAC). The goal of these experiments was to get a general sense of mass transfer rates and adsorptive capacities when treating NSF 350 gray water. Representative data sets for reduction of COD values in gray water as a function of time and flux in upflow GAC columns are presented in Figure 1. For the range of loading conditions tested, lower flux values resulted in greater reductions in COD values. Surfactant removal, as measured by reduction in foam formation level (FFL), was in excess of the 90% (effluent FFL below limit of detection).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Unit of measure</th>
<th>Average value</th>
<th>95% CI</th>
<th>n</th>
<th>NSF 350 recommended range</th>
</tr>
</thead>
<tbody>
<tr>
<td>BOD-5</td>
<td>mg/L</td>
<td>176 ±20</td>
<td>7</td>
<td>130–180</td>
<td></td>
</tr>
<tr>
<td>FFL</td>
<td>mm</td>
<td>13 ±1</td>
<td>17</td>
<td>NA</td>
<td></td>
</tr>
<tr>
<td>COD</td>
<td>mg/L</td>
<td>392 ±28</td>
<td>26</td>
<td>250–400</td>
<td></td>
</tr>
<tr>
<td>TSS</td>
<td>mg/L</td>
<td>115 ±15</td>
<td>5</td>
<td>80–160</td>
<td></td>
</tr>
<tr>
<td>Turbidity</td>
<td>NTU</td>
<td>58 ±12</td>
<td>12</td>
<td>50–100</td>
<td></td>
</tr>
<tr>
<td>Phosphorous</td>
<td>mg P/L</td>
<td>6.7 ±1.0</td>
<td>5</td>
<td>1.0–3.0</td>
<td></td>
</tr>
<tr>
<td>TKN</td>
<td>mg N/L</td>
<td>2.4 ±0.3</td>
<td>5</td>
<td>3.0–5.0</td>
<td></td>
</tr>
<tr>
<td>pH</td>
<td>pH units</td>
<td>7.5 ±0.2</td>
<td>17</td>
<td>6.5–8.0</td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>°C</td>
<td>23 ±2</td>
<td>11</td>
<td>25–35</td>
<td></td>
</tr>
</tbody>
</table>

CI, confidence interval.
for the lower two flux conditions, but decreased to 75% at the high flux condition after 300 minutes. Turbidity reduction levels decreased with increasing flux, with only 30% reduction achieved after 300 minutes at the highest flux condition. Contaminant removal levels were generally higher at shorter operating times and lower flux values. Of the parameters measured, FFL appeared to be reduced most efficiently by the GAC media, confirming that gray water surfactants are removed relatively efficiently from gray water by GAC. For all parameters, contaminant breakthrough increased over relatively short operating times and low flux values, confirming that adsorption alone would have limited sustained capacity in treating real gray water without frequent regeneration of the GAC.

While chemical or thermal regeneration could be considered for restoring the adsorptive capacity of GAC, those methods would add complexity and cost to a gray water treatment process and not be feasible in distributed design environments. Therefore, the potential benefit of biodegradation within a GAC column was explored. To this end, continuous-loading BAC filtration experiments were performed. The BAC filtration experiments were performed for longer periods of time to assess bioregenerability and with water flux values lower than those shown in Figure 1 to account for slower mass transfer that may result from biofilm-coated media. Results of the continuously operated BAC filter test are shown as one of the data sets in Figure 2.

At a flux value of 1.3 Lpm/m², an average COD reduction of 74% was observed throughout the duration of the 5-week period, though the maturation phase (day 1–4) was not fully characterized. The results indicate that equilibrium was achieved between mass transfer, sorption, biodegradation, and biomass removal, resulting in sustained removal of organics without the use of chemical or thermal regeneration. The BAC filter required washing to remove biofilm for 5 minutes every 2 days in order to sustain performance. However, while sustained removal of organics was achieved, effluent from the continuous BAC filter was odorous, based on human observation during experimentation, due to anoxic and/or anaerobic conditions within the continuously operated biofilters.

As an alternative to conventional continuously operated BAC filtration, a newly developed IOBAC was assessed. In the IOBAC approach, aerobic conditions are promoted through the application of intermittent loadings at higher rates followed by draining and idling of the filter for sustained periods to facilitate biodegradation. This new approach could also accommodate intermittent loading schedules common in gray water systems, and it reduces odor-generating anoxic and/or anaerobic biodegradation activity within the media bed. The IOBAC filter was tested...
at a three-fold higher loading rate (4 Lpm/m²) for only 33% of each day, thus maintaining a volumetric loading rate equivalent to that used in the BAC studies. The resulting reduction of COD levels in gray water by IOBAC filtration is shown in Figure 2. After an initial maturation phase, the IOBAC filter provided a consistent COD removal of approximately 55–60%, based on grab samples collected 1 hour and 3 hours into the semi-daily 4-h loading cycles. In general, the data indicated that the IOBAC performance decreases slowly within each 4-h run period but recovers prior to the next run due to the bioregeneration effect. Consistent with the BAC test, the IOBAC results indicate that equilibrium was achieved between mass transfer, sorption, biodegradation, and biomass removal, resulting in sustained removal of organics. It was noted during testing and optimization that more frequent washing (once every 2 days) was more effective than less frequent washing (twice each week) in providing sustained performance. This performance was sustained over a period of 6 months of weekly measurements (data after 2 months not shown). While the average effluent COD level was not as low as that from the BAC filter, the effluent from the IOBAC filter was not noticeably odoriferous, based on human observation during the experiments.

For both BAC and IOBAC approaches, the effluent COD values were higher than desired for many water reuse applications. Therefore, their augmentation with potentially complementary technologies was investigated. Figure 2 presents a good example of this approach, showing that pretreatment of gray water with polymeric coagulant upstream of biofiltration can improve performance. By mixing 5 mg/L of PDM into the feed tank, effluent COD levels generally decreased another 20–25%, yielding sustained improvements of product water with COD values of less than 40 mg/L for IOBAC filters that had been running for over 4 months. This enhancement is likely due to an enhanced removal of small organic-laden particulate matter due to charge neutralization as well as flocculation within the feed tank and filter, resulting in improved filter efficiency. Further efficiency gains were realized by increasing the filter column height while maintaining equivalent volumetric loading rates by increasing the flux by the ratio of new to old filter depth. In the optimized IOBAC system, augmented with polymeric coagulant pretreatment, COD value reductions in excess of 90% were observed. A similar result was reported previously (Sostar-Turk et al. 2005), but that study used metal salt coagulants followed by a settling step and was tested for a relatively short period. The use of biodegradable polymeric coagulant in the present study may lead to better sustained performance.

The potential benefit of applying coagulation and sedimentation alone was also considered in this study. Previous studies have explored the utility of conventional metal salt coagulants with some success (Cui & Ren 2005; Pidou et al. 2008). However, metal salts such as alum and ferric chloride may not be ideal for gray water treatment as they require high mass concentrations, can contribute to scale or foulant formation if not applied properly, and can be difficult or hazardous to handle in some forms. In the present study, the use of the cationic organic polymer PDM was investigated because it can be applied at lower doses than conventional metal salt coagulants and can be biodegraded. Jar tests were performed to identify optimal dosing for charge neutralization. Results of jar tests performed with gray water and increasing doses of PDM coagulant (0.5, 1, 5, 10, 50, and 100 mg/L) are shown in Table 3. Floc formation was visually apparent at doses as low as 5 mg/L, and turbidity and COD level reductions improved with increasing coagulant dosage up to maximum values at 50 mg/L of PDM. This trend was also consistent with visible observation of larger flocs being formed as the dose increased from 5 to 50 mg/L. However, at a PDM coagulant dosage of 100 mg/L, the turbidity and COD increased relative to the untreated control, yielding negative (−) removals, consistent with a charge re-stabilization mechanism, and COD values actually increased due to the coagulant. Additionally, for all conditions, the flocs formed

<table>
<thead>
<tr>
<th>Coagulant dosage (mg/L)</th>
<th>Turbidity removal (%)</th>
<th>COD removal (%)</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.5</td>
<td>2 ± 1</td>
<td>−0.45 ± 1.2</td>
<td>3</td>
</tr>
<tr>
<td>1</td>
<td>3 ± 0.44</td>
<td>2 ± 1</td>
<td>3</td>
</tr>
<tr>
<td>5</td>
<td>66 ± 5</td>
<td>25 ± 3</td>
<td>3</td>
</tr>
<tr>
<td>10</td>
<td>86 ± 3</td>
<td>35 ± 6</td>
<td>3</td>
</tr>
<tr>
<td>50</td>
<td>91 ± 6</td>
<td>40 ± 2</td>
<td>3</td>
</tr>
<tr>
<td>100</td>
<td>−157 ± 51</td>
<td>−8 ± 5</td>
<td>3</td>
</tr>
</tbody>
</table>
were generally difficult to settle. However, given the results indicating that charge neutralization begins at low coagulant doses, PDM could still be beneficial for gray water treatment when combined with other treatments, as shown with the IOBAC tests (Figure 2).

The use of ion exchange for gray water treatment was also studied as a potential complementary technology to BAC or IOBAC filters, given that ion exchange resins employ a charge-mediated removal mechanism that is distinct from GAC adsorption which primarily leverages non-ionic forces. IX resin can also be readily regenerated in many applications using brine solution, and it has a long track record in common use within households for hardness control. In the present study, Tanex resin was chosen for evaluation based on its selectivity for dissolved organics such as tannins. To protect the resin from colloidal fouling, per manufacturer recommendations, the gray water was first ultrafiltered to remove particulates, which reduced the turbidity and COD of the challenge water by approximately 99 and 50%, respectively. The distribution of organic contaminant types within the challenge water was also likely altered. Using this modified challenge water, results for the breakthrough of the organics in the Tanex column over multiple loading cycles are shown in Figure 3. COD breakthrough profiles appeared to be biphasic, with a very fast initial increase in the fraction of organics breaking through the column, followed by a long and not fully characterized phase of slower breakthrough. After an initial rapid phase of breakthrough, a sustained COD removal of approximately 60% was maintained, even for the longest run times tested. This sustained removal may have been ascribed to a non-IX removal mechanism. Regeneration with 10% sodium chloride appeared to restore performance to baseline levels over the five runs conducted. One sample showed high initial breakthrough (Run 2), but this was ascribed to incomplete removal of the regenerant salt solution prior to starting the next run (Run 3). Additional variability in the profiles may be partly attributable to variations in the source water, since the experiments were performed over the course of several weeks. In general, IX treatment as tested herein was only moderately effective against a pretreated gray water source and was considerably more logistically intensive in terms of consumable material requirements for regeneration.

The use of MF and UF membrane technology for the treatment of gray water was also investigated in this study. A summary of results is provided in Table 4. As expected, decreasing the pore size resulted in higher levels of purification, though diminishing returns were observed in the ultrafiltration range. A polypropylene cartridge filter provided little change in water quality compared to controls. MF reduced the levels of turbidity substantially and also had some impact on COD and FFL values. UF membranes provided 55–60% COD removal, as well as very high levels of turbidity removal (>99%). These results are generally consistent with previous studies with other low-pressure membrane systems (Ramon et al. 2004; Li et al. 2008; Hocaoglu & Orhon 2013). However, one notable result

![Figure 3](https://iwaponline.com/jwrd/article-pdf/5/3/239/377382/jwrd0050239.pdf)

**Table 4** Performance of low-pressure (30 psi) membrane filters with varying pore size values for the treatment of gray water

<table>
<thead>
<tr>
<th>Membrane type</th>
<th>Pore size</th>
<th>COD removal (%)</th>
<th>FFL reduction (%)</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP cartridge filter</td>
<td>10 μm</td>
<td>4 ± 1</td>
<td>0</td>
<td>3</td>
</tr>
<tr>
<td>PES microfilter</td>
<td>0.2 μm</td>
<td>23 ± 9</td>
<td>2 ± 5</td>
<td>4</td>
</tr>
<tr>
<td>PES ultrafilter</td>
<td>0.05 μm</td>
<td>56 ± 7</td>
<td>−74 ± 30</td>
<td>5</td>
</tr>
<tr>
<td>PVDF ultrafilter</td>
<td>0.01 μm</td>
<td>59 ± 5</td>
<td>−48 ± 17</td>
<td>3</td>
</tr>
</tbody>
</table>

PP, polypropylene; PES, polyethersulfone; PVDF, polyvinylidine fluoride.
from the present study was that FFL generally increased relative to untreated controls, despite decreasing the COD value of the sample. Thus, UF was found to yield negative (−) FFL reduction values. The higher FFL observed in the effluent may be due to the reduced availability of particulate surface area for surfactant uptake, which might indicate that particle-associated surfactants are still able to permeate the membrane by dissolving out of the fouling layer and into the permeate. Even with the increase in FFL and only partial COD level decrease observed, UF treatment appears to be a robust treatment technology for gray water treatment in conjunction with BAC or IOBAC filtration, which can remove surfactants efficiently, due to the very high level of turbidity reduction.

Given the diverse composition of gray water and results from individual technology evaluations, it was clear that combinations of the component technologies are needed to remove a broader range of contaminants. To this end, multi-barrier arrangements with expected additive performance were assessed. Table 5 summarizes the performance impacts of the multi-barrier approaches investigated. The most effective binary combination tested for removal of organics was the use of a 5 mg/L dose of coagulant followed by IOBAC filtration (Coag + IOBAC), which resulted in 84% decrease in COD. For particulate removal, any systems that incorporated UF produced a 99.5% decrease in turbidity. The most effective ternary system tested was coagulation, IOBAC, and UF (Coag + IOBAC + UF), which resulted in a 94% decrease in COD and 99.5% decrease in turbidity.

To explore sustained operation of this ternary system in greater detail, particularly with respect to membrane performance, further studies of combined treatments were performed to assess the flux decline and recovery. The results of the UF flux studies are provided in Figure 4. For each condition, three runs were conducted on a single membrane, with an extensive clean-water backflush cycle applied between each run. Both the raw and IOBAC-pretreated gray waters resulted in flux declines of nearly 70% within 40 minutes, indicating that the removal of organics by IOBAC alone did not effectively remove any of the fouling portions of the gray water associated with flux decline. Furthermore, compared to raw gray water, the IOBAC-treated water exhibited reduced flux recovery after backflushing, potentially due to removal of the surfactants in the raw gray water and a decreased ability to dislodge pore-clogging colloids. However, the addition of a coagulant upstream of the IOBAC filter reduced flux declines to 40% and restored flux recovery capability. This result was not observed when adding coagulant alone without the IOBAC step, though coagulant addition did appear to assist in slowing the extent of flux decline observed.

Overall, the results obtained in this research supported the objective to assess BAC filtration and complementary technologies for the control of particulate and organic contaminants in gray water. The evaluation of several different technologies using a consistent synthetic gray water formulation provided a good basis for comparison and component technology integration. IOBAC filtration was identified as a new operation strategy for sustained removal of organics while limiting energy requirements and odor formation through passive aeration. While individual performance of the component technologies studied was generally insufficient for many water reuse activities, the additive benefit of combining complementary physicochemical and biological treatments yielded much better water quality. Optimal combinations, such as coagulation followed by IOBAC filtration and UF, produced relatively high quality product water with low turbidity and reduced organic content. Under the conditions tested, COD and turbidity reduction levels of more than 90 and 99% were achieved.

<table>
<thead>
<tr>
<th>Technology</th>
<th>Turbidity removal (%)</th>
<th>COD removal (%)</th>
<th>FFL reduction (%)</th>
<th>n</th>
</tr>
</thead>
<tbody>
<tr>
<td>IOBAC only</td>
<td>35</td>
<td>65</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>MF only</td>
<td>95</td>
<td>10</td>
<td>0</td>
<td>4</td>
</tr>
<tr>
<td>UF only</td>
<td>99.5</td>
<td>58</td>
<td>−60</td>
<td>7</td>
</tr>
<tr>
<td>Coag. + IOBAC</td>
<td>78</td>
<td>84</td>
<td>100</td>
<td>6</td>
</tr>
<tr>
<td>IOBAC + MF</td>
<td>95</td>
<td>69</td>
<td>100</td>
<td>3</td>
</tr>
<tr>
<td>IOBAC + UF</td>
<td>99.5</td>
<td>75</td>
<td>100</td>
<td>3</td>
</tr>
<tr>
<td>UF + IX</td>
<td>99.5</td>
<td>82</td>
<td>55</td>
<td>5</td>
</tr>
<tr>
<td>IOBAC + UF + IX</td>
<td>99.5</td>
<td>88</td>
<td>100</td>
<td>2</td>
</tr>
<tr>
<td>Coag + IOBAC + UF</td>
<td>99.5</td>
<td>94</td>
<td>100</td>
<td>4</td>
</tr>
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respectively. These additive performance effects are consistent with observations in the component technology evaluations: i.e., IOBAC was highly effective against surfactants that were not efficiently removed by UF membranes, based on relative FFL of the effluents.

Many treatment technologies spanning physical, chemical, and/or biological processes have been previously studied for gray water treatment (Winward et al. 2008; Li et al. 2009; Ghunmi et al. 2011). However, due to variability in source water characteristics, treatment methods, and analytical methods used across previously published studies, it is often difficult to confidently compare the wide range of gray water treatment technologies. Additionally, many previous studies were performed on a focused set of technologies and for limited time periods that may not fully represent equilibrated operating conditions, whereas sustained performance of a wide range of technologies was a key focus of the present study. Nevertheless, the beneficial effect of combining different types of treatment processes in a multi-barrier system noted herein is consistent with the literature. Li et al. (2009) suggested that while physical processes alone often fail to adequately remove organics and other soluble constituents, the combination of physical filtration and aerobic biological treatment was the most feasible treatment for medium to high strength gray water recycling. Ghunmi et al. (2011) similarly concluded that purely physical systems should be avoided due to the excess non-stabilized sludge production. Strictly biological gray water treatment systems also have challenges with respect to needs for continuous flow, operational support, and physical space. Previous biological gray water treatment studies include biofilters (Jefferson et al. 2001; Hu et al. 2005; Gross et al. 2007), aerobic rotating biological contactors (Nolde 1999; Friedler et al. 2005; Friedler et al. 2006), anaerobic bioreactors (Elmitwalli & Otterpohl 2007; Hernandez et al. 2008), and membrane bioreactors (Liu et al. 2005; Lesjean & Gniirs 2006; Merz et al. 2007; Winward et al. 2008). The membrane bioreactor may be the most promising of the previous technologies studied, as it includes a combined biological and physical approach.

In the updated USEPA Guidelines for Water Reuse (USEPA 2012), the recommended water quality limits for unrestricted reuse are 30 mg/L BOD, 5 NTU, and 100 colony forming units (cfu)/100 ml fecal coliform bacteria. For unrestricted, non-potable urban reuse, the recommended limits for BOD, turbidity, and fecal coliform bacteria are 10 mg/L, 2 NTU, and 1 cfu/100 ml, respectively. These values are generally consistent with the NSF/ANSI Standard 350 (NSF International 2011) performance metrics. The IOBAC process and supporting technologies studied herein could also offer a robust alternative approach for the design of systems for restricted and unrestricted water reuse activities. Further integration with a disinfection system would be needed. While UF treatment alone would likely provide substantial pathogen reduction credits, residual disinfection would be required for most applications (USEPA 2012).

The PDM/IOBAC/UF approach may also be beneficial for higher tier reuse applications. Aside from guidance for Army field operations generated by the United States Army.
Public Health Command (Fifty 2008), there is limited guidance available in the USA for higher tier reuse applications such as showering. In the Army, near-potable levels of purification are practiced via incorporation of reverse osmosis (RO) polishing of product water. Achieving near-potable water quality levels for reuse applications, like showering, while maximizing robustness and energy efficiency in a distributed design application presents a continued design challenge. Further studies on emerging contaminants like pharmaceuticals, personal care products, and other micropollutants may drive future designs and new requirements. For instance, removal of organic disinfection by-product (DBP) precursors upstream of chlorination processes may be desirable for showering which represents a primary route of exposure to DBPs (Backer et al. 2000), and given that DBP formation at levels above Environmental Protection Agency maximum contaminant limits has been reported for reclaimed water sources subjected to chlorination (Chen & Westerhoff 2010) or other disinfection treatments (Beck et al. 2015). In addressing these challenges, technologies such as PDM/IOBAC/UF could be used for pre-treatment upstream of advanced purification technologies, such as RO membranes and/or advanced oxidation systems. Such pretreatment could reduce organic loadings, provide multi-barrier protection, and potentially yield higher levels of purity. From an operational perspective, previous bench scale studies have indicated that biological and physiochemical pretreatment of gray water can reduce fouling of RO membranes (Crawley et al. 2012), though these effects would potentially be scenario specific.

CONCLUSIONS

This study compared the performance of various water treatment methods for the removal of organic and particulate contaminants from gray water. Based on the results, the following conclusions are made:

- BAC filtration provided sustained removal of organics from gray water. A new IOBAC, in which the BAC filter is periodically taken off-line and drained to facilitate passive aeration, still provided sustained performance after months of operation. The IOBAC approach may be well suited for managing intermittent flows, and it may also reduce odor of the product water by promoting aerobic bioregeneration.
- A combination of polymeric coagulant, IOBAC filtration, and UF exhibited an additive treatment effect by targeting complementary contaminant fractions of the gray water matrix, reducing turbidity and COD levels by 99.5 and 94%, respectively. Therefore, this strategic combination of technologies could support reuse applications ranging from irrigation to toilet flushing. To support higher tier reuse applications such as showering, the technologies studied herein could be considered for efficient pretreatment ahead of advanced purification technologies such as RO.

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First received 7 October 2014; accepted in revised form 10 January 2015. Available online 26 March 2015