Potable reuse of wastewater effluent through an integrated soil aquifer treatment (SAT) – membrane system

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Abstract Soil aquifer treatment (SAT) has been shown to provide significant reductions of wastewater effluent organic matter (EfOM). While SAT can renovate wastewater effluent to levels commensurate with potable reuse, SAT represents only a single treatment barrier. Membrane filtration has been demonstrated to be an alternative treatment process for wastewater reclamation. However, the performance of membrane filtration is significantly constrained by membrane fouling, reducing membrane productivity in terms of flux decline. Soil aquifer treatment (SAT) is used as a primary barrier in the reclamation system to remove a complex suite of chemical and biological contaminants in wastewater. This study of membrane filtration of SAT-source waters is intended to determine the benefits of the SAT system in terms of membrane fouling reduction with membrane filtration providing a secondary barrier. Membrane treatment of SAT-recovered water provides a multiple barrier exhibiting additional organic matter removal. Moreover, the SAT system also provides for storage of reclaimed water. The synergy between SAT and membrane treatment has led to the concept of an integrated SAT-membrane system for potable reuse.

Keywords Membrane fouling; nanofiltration; potable reuse; soil aquifer treatment (SAT); ultrafiltration

Introduction

In recent research, it has been found that soil aquifer treatment (SAT) can renovate wastewater secondary effluent to levels commensurate with potable reuse (i.e. drinking water quality). A SAT system, consisting of a recharge basin, unsaturated (vadose) zone infiltration, saturated zone mixing and groundwater transport, and extraction by a recovery well; has been shown to provide significant reductions of wastewater effluent organic matter (EfOM), viruses, and nitrogen (ammonia and nitrate). A research study at two field sites in Arizona has demonstrated dissolved organic carbon (DOC) reductions from 5–15 mg/L to ~1 mg/L, multiple-log reductions of bacteriophage (a surrogate of enteric viruses), and effective nitrogen removal through subsurface transformations including nitrification and subsequent denitrification. However, from a regulatory perspective, SAT represents only a single treatment barrier. Presently, in the USA, approval of a potable reuse project would require multiple treatment barriers within the context of an indirect potable reuse scheme. Membrane treatment of SAT-recovered water provides a multiple barrier approach for viruses while providing additional organic matter removal. The membrane component can be either ultrafiltration (UF) to provide additional log-removal of viruses, or nanofiltration (NF) to provide removals of viruses, organic matter, and potentially other contaminants such as endocrine disrupting compounds (EDCs) and pharmaceutical-active compounds (PhACs). SAT has been found to be an effective pretreatment for membranes, providing a significant reduction in membrane fouling. This synergy between SAT and membrane treatment has led to the concept of an integrated SAT-membrane system for potable reuse.

This study performed parallel membrane filtration experiments with feed waters corresponding to secondary effluents and/or tertiary effluents (prior to introduction into a...
recharge basin) and post SAT reclaimed waters extracted after both unsaturated and saturated zone treatments. The purpose of the study was to evaluate the performance of SAT, membrane filtration, and an integrated SAT-membrane system in terms of organic removal and reduction in membrane fouling potential.

**Materials and methods**

**Membranes**

Two different membranes, including an NF membrane (ESNA) and a UF membrane (GM), were employed. The properties of each membrane are shown in Table 1. The ESNA and GM membranes are made of polyamide (PA). The ESNA membrane is considered to be an NF membrane, based on its nominal molecular weight cut-off (MWCO), while the GM membrane is considered to be a tight UF membrane. The contact angle of each membrane characterizes the hydrophobicity of the membrane surface. The greater the contact angle, the greater the hydrophobicity. The ESNA exhibited a relatively large hydrophobicity. Surface charges of the membranes, determined by zeta potential, help explain electrostatic interaction between membranes and solutes and/or foulants. The GM membrane exhibited a high negative surface charge in terms of zeta potential.

Additionally, clean and fouled membranes were analyzed by attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR) to determine functional groups associated with membrane surface materials and foulants.

**Membrane filtration experiments**

Flux decline tests were performed with a dead-end stirred cell membrane unit (Amicon, USA). The system consists of a stirred cell membrane unit, a 4 litre feed reservoir, and a magnetic stirrer. The membrane unit, accommodating an active area for permeation of 28.7 cm², employs 62 mm disc membrane specimens. The feed reservoir provides a continuous supply of feed water to the stirred cell membrane unit. A magnetic stirrer within the unit is operated at a speed of approximately 300 rpm to provide a crossflow velocity. Nitrogen gas is used to apply the pressure to the membrane unit. The system is operated in a constant pressure (declining flux) mode, and at room temperature (~20°C).

A membrane specimen is soaked in Milli-Q water for a day before the test to remove soluble materials coating the membrane surface. Milli-Q water is then fed through the membrane. The permeate flux of Milli-Q water is monitored until it becomes stable, then pure water permeability (PWP) is measured. The initial flowrate of Milli-Q water is adjusted to between 1.5 and 1.8 mL/min by adjusting the transmembrane pressure. Two litres of source water are then placed into the feed reservoir. Aliquots of 200 mL of permeate are collected up to 1,600 mL of total permeate volume. The permeate flowrate, and DOC and UVA₂₅₄ of permeate samples, are monitored as a function of permeate volume. Additionally, the final retentate within the stirred cell is collected to measure DOC and UVA₂₅₄, indicating EfOM rejections.

**Table 1** Characteristics of membranes

<table>
<thead>
<tr>
<th>Membrane</th>
<th>Type</th>
<th>Material</th>
<th>MWCO</th>
<th>Contact angle (°)</th>
<th>Zeta potential (mV) at pH 7*</th>
<th>Pure water permeability (L/m²•day•kPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ESNA</td>
<td>NF</td>
<td>PA</td>
<td>200</td>
<td>60.3</td>
<td>-11.5</td>
<td>1.35</td>
</tr>
<tr>
<td>GM</td>
<td>UF</td>
<td>PA</td>
<td>8,000</td>
<td>45.5</td>
<td>-17.0</td>
<td>2.96</td>
</tr>
</tbody>
</table>

*Cho et al. (1998)*
Source (feed) waters

Different source (feed) waters from two operating SAT facilities, the Mesa Northwest Water Reclamation Plant (NWWRP) in Mesa (Arizona, USA) and the Sweetwater Underground Storage and Recovery Facility (SUSRF) in Tucson (Arizona, USA), were used in membrane filtration tests. These source (feed) waters, consisting of Mesa tertiary effluent (ME-TE) derived from secondary biological treatment followed by tertiary sand filtration, and Tucson secondary effluent (TU-SE), were used as sources of reclaimed water. They have been subjected to further treatment by percolation through the soil mantle prior to mixing with groundwater, i.e. SAT. The SAT treated waters, including OW2-90 (Mesa), SIBW-6U (Mesa), and WR199A (Tucson) were collected from groundwater recovery wells, located down gradient from the infiltration basins. The OW2-90 sample was extracted from the OW2 well, approximately 700 feet down gradient from the infiltration basin. The travel time was estimated to be approximately 6 months. The SIBW-6U sample was collected from the SIBW-6U extraction well. The SIBW-6U well was located 1 mile distant from the recharge basin with an estimated travel time of approximately 6 years. The WR199A sample was extracted from the groundwater, typically about 120 feet below ground surface. The WR199A well is located adjacent to the recharge basin. The travel time was approximately 2 months. These SAT treated waters were evaluated in the membrane filtration tests.

All source waters were 0.45-µm filtered and stored at 4°C until analysis. Dissolved organic carbon (DOC) and ultraviolet absorbance (UVA) at a wavelength of 254 nm were analyzed using a total organic carbon (TOC) analyzer (TOC 800, Sievers) and UV-visible spectrophotometer (UV160A, Shimadzu), respectively. Specific UVA (SUVA), a ratio of UVA and DOC, represents an index of aromaticity. Conductivity and total dissolved solids (TDS) were measured by a conductivity meter (Orion115). The characteristics of each source (feed) water are shown in Table 2.

High performance liquid chromatography-size exclusion chromatography (HPSEC) with on-line UVA and DOC detectors was used to characterize molecular mass distribution of wastewater effluent organic matter (EfOM). A highly buffered sodium sulfate was used as a mobile phase. The column media in HPSEC is porous gel, which allows a separation of macromolecules at high flowrates. Smaller molecules can access more of the internal pore volume than larger molecules. The net result is that large molecules elute first followed by progressively smaller molecules (Her et al., 1999). The response over separation time was measured using a Shimadzu SPD-6A UV spectrophotometer and a Sievers 800 total organic carbon (TOC) analyzer. The magnitude of response detected in the chromatogram peaks is a relative response that allows qualitative comparison between corresponding samples. This technique provides an understanding of the fate of organic compounds during SAT and the rejection of organic matter by membrane filtration, and provides insight to potential organic foulants. The results help determine the performance of the SAT system, membrane filtration, as well as an integrated system of both alternative technologies.

Table 2 Characteristics of source (feed) waters

<table>
<thead>
<tr>
<th>Source water</th>
<th>DOC (mg/L)</th>
<th>UVA_{254} (1/cm)</th>
<th>SUVA (L/mg-m)</th>
<th>pH (-)</th>
<th>Conductivity (µS/cm)</th>
<th>TDS (mg/L)</th>
<th>% reclaimed water</th>
</tr>
</thead>
<tbody>
<tr>
<td>NWWRP (Mesa)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ME-TE</td>
<td>5.71</td>
<td>0.086</td>
<td>1.51</td>
<td>8.34</td>
<td>1,269</td>
<td>608</td>
<td>n/a</td>
</tr>
<tr>
<td>OW2-90</td>
<td>1.12</td>
<td>0.020</td>
<td>1.79</td>
<td>8.53</td>
<td>1,340</td>
<td>643</td>
<td>95</td>
</tr>
<tr>
<td>SIBW-6U</td>
<td>0.97</td>
<td>0.023</td>
<td>2.37</td>
<td>8.42</td>
<td>1,150</td>
<td>550</td>
<td>75</td>
</tr>
<tr>
<td>SUSRF (Tucson)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>TU-SE</td>
<td>13.59</td>
<td>0.161</td>
<td>1.19</td>
<td>7.20</td>
<td>732</td>
<td>342</td>
<td>n/a</td>
</tr>
<tr>
<td>WR199A</td>
<td>0.83</td>
<td>0.018</td>
<td>2.17</td>
<td>8.31</td>
<td>790</td>
<td>379</td>
<td>100</td>
</tr>
</tbody>
</table>
Results and discussion

Organic removal by soil aquifer treatment (SAT)

SAT has been found to provide effective removal of bulk organic matter (EfOM) present in secondary or tertiary effluents. According to Table 2, the DOC of ME-TE and TU-SE has been reduced during SAT. At NWWRP (Mesa), approximately 80 per cent of DOC was removed during percolation through the soil mantle. An additional few per cent DOC removal was observed in the down gradient flow to the SIBW-6U well, although there is also the influence of some dilution affecting samples extracted from the SIBW-6U well. This result reflects the biorefractory character of organic compounds after SAT. At SUSRF (Tucson), the reduction of DOC during SAT was found to be as high as 90 per cent.

The SEC-DOC chromatograms of ME-TE compared to those of OW2-90 and SIBW-6U are shown in Figure 1. The result indicates the transformation of organic matter during SAT. Large molecules with no UV-absorbance, eluting between 1,300 and 1,900 seconds and corresponding to polysaccharide-like compounds, are effectively removed during percolation through the soil. Some humic substances, reflected by high DOC and high UV-absorbance peaks, as well as some low molecular mass compounds with low UV-absorbance are removed during SAT. However, humic substances, eluting between 2,200 and 3,500 seconds and including humic acids and fulvic acids, still remain in the SAT treated water, with SAT treated waters exhibiting a high SUVA and displaying a refractory behavior. The chromatograms also shows the peak, eluting at 4,000 seconds, with low UV-absorbance; this peak corresponds to low-molecular weight organic acids which may be transitory by-products of biodegradation.

The most interesting observation from the SEC-DOC chromatograms of the NWWRP (Mesa) series is the similarities observed between the OW2-90 sample and the down gradient SIBW-6U sample. This observation indicates that minimal additional treatment is afforded by the added years of storage in the aquifer and further suggests that the majority of the remaining DOC after six months of treatment to the OW2 well is a persistent fraction. These results are consistent with the per cent removals of DOC achieved by OW2-90 and SIBW-6U, which are 80 and 83%, respectively. Figure 2 illustrates the SEC-DOC chromatogram of TU-SE and WR199A. The results show similar trends as mentioned above.

Membrane filtration and flux decline

During membrane filtration, a decrease in permeate flux reflects an increase of the mass of materials deposited on/into the membrane surface/pores. Figure 3 illustrates flux decline trends of Mesa tertiary effluent (ME-TE), and corresponding SAT treated effluents from...
OW2-90 and SIBW-6U wells. The OW2-well is located approximately 700 feet downgradient from the recharge basin and the sample from this well contains approximately 95 per cent reclaimed water. The SIBW-6U well is located about 1 mile downgradient from the recharge basin and the sample from this well contains 75 per cent reclaimed water. Flux decline tests were conducted using the ESNA and GM membranes. The results show the different flux decline trends among the various source (feed) waters. The ME-TE exhibits higher (normalized) flux decline than the others, implying that organic and colloidal fouling potential is reduced by SAT. However, with the GM membrane, there is no significant difference in flux decline trends between ME-TE and corresponding SAT-treated waters.

The flux decline trends of Tucson secondary effluent (TU-SE), compared to that of SAT treated water from WR199A well, are shown in Figure 4 for the ESNA and GM membranes. The WR199A well is located adjacent to the recharged basin and the per cent reclaimed water is estimated as 100%. These results indicate that the TU-SE shows higher (normalized) flux decline than the WR199A. The permeate flux is affected by the high effluent organic matter (EfOM) contained in the reclaimed water source. The reduction in permeate flux of WR199A is lower than that of TU-SE, indicating that EfOM is removed during transport through the soil.

The average rejections by NF and UF membranes based on DOC and UVA are shown in Table 3. The significant mechanisms of EfOM rejection are size exclusion (MWCO) and charge interaction (electrostatic repulsion), whereas EfOM-fouling of NF and UF membranes is affected by MWCO and hydrophobic interactions (Cho et al., 1998). The ESNA membrane was found to be very effective in rejection of EfOM and inorganic compounds.
due to its MWCO and surface charge. Electrostatic effects were likely dominant in organic rejection by the GM membrane.

The ESNA and GM membranes showed high organic matter rejections for wastewater effluents, i.e. ME-TE and TU-SE. However, these source waters caused high flux decline, leading to significant reductions in membrane productivity. Flux decline results showed that the SAT-treated waters exhibit significantly less flux decline, reflecting the effective performance of an integrated SAT-membrane system. Table 3 also compares the performance of membranes, SAT as an alternative technology, and the integrated SAT-membrane system.

**FTIR spectra of clean and fouled membranes**
Attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR) was used to analyze for functional groups of foulants on membrane surfaces. Some of the peaks of clean membranes were changed in absorbance intensity when the membranes were fouled, illustrating foulant functional groups. The differences in IR spectra between clean and fouled membranes may explain the adsorption phenomena of the foulants. Figure 5 illustrates the comparison of IR peaks between clean ESNA and GM membranes and corresponding fouled membranes with ME-TE and SAT treated waters, including OW2-90 and SIBW-6U. According to Leenheer et al. (2000), the peaks at wave numbers of 1,540 cm\(^{-1}\) and/or 1,640 cm\(^{-1}\) indicate the functional groups of primary and secondary amides. The peaks between 1,040 and 1,240 cm\(^{-1}\) show C-O bonds of ethers, carboxylic acids, and polysaccharides (Cho et al., 1998). Figure 6 shows the differences of IR peaks between TU-SE and WR199A for ESNA and GM membranes. The difference in absorbance intensity of IR peaks around 960–1,170 cm\(^{-1}\) indicates the possibility of polysaccharides and/or polysaccharide-like substances as foulants. The IR peaks around 1,540 cm\(^{-1}\) and 1,640 cm\(^{-1}\) indicate the adsorption of proteins and/or protein-like materials contained in wastewater EfOM. The IR peaks of ME-TE and TU-SE were found at 1,040, 1,540, and 1,640 cm\(^{-1}\), indicating the possible foulants as polysaccharides, proteins, and/or amino sugars. These possible foulants were found to be reduced in the SAT-treated waters, reflecting the effectiveness of SAT as a membrane pretreatment.

**SEC-DOC chromatograms of feed and permeate waters**
The SEC-DOC chromatograms of ME-TE feedwater and ME-TE after membrane filtration using ESNA and GM membranes are shown in Figure 7; those of TU-SE feedwater and TU-SE permeate from ESNA and GM membranes are shown in Figure 8. The comparisons of SEC-DOC chromatograms between feedwater and permeate illustrate the high efficiency of the ESNA membrane in the removal of organic compounds contained in reclaimed water. However, the performance of the GM membrane is significantly lower in terms of organic matter rejection.

### Table 3  The performance of ESNA and GM membranes vs. SAT vs. integrated SAT-membrane system

<table>
<thead>
<tr>
<th>Source water</th>
<th>DOC rejection (%)</th>
<th>UVA rejection (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NWWRP (Mesa)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>ME-TE</td>
<td>94</td>
<td>92</td>
</tr>
<tr>
<td>OW2-90</td>
<td>82</td>
<td>90</td>
</tr>
<tr>
<td>SIBW-6U</td>
<td>81</td>
<td>83</td>
</tr>
<tr>
<td>SUSRF (Tucson)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TU-SE</td>
<td>97</td>
<td>95</td>
</tr>
<tr>
<td>WR199A</td>
<td>73</td>
<td>89</td>
</tr>
</tbody>
</table>

The SAT renovated waters were also used as feedwaters in membrane filtration. Both feed and permeate waters were characterized by SEC analysis. Figures 9 and 10 show the SEC-DOC chromatograms of OW2-90 and SIBW-6U samples, respectively, using ESNA and GM membranes. The results indicate complete rejection of organic compounds by the integrated SAT-NF membrane system, whereas most organic compounds were still remaining in the UF permeate. The decision of whether to select UF or NF after SAT is based on a number of factors, including system productivity (i.e. higher specific flux for UF), susceptibility to fouling (higher for NF), treatment objectives (viruses by UF; viruses, organic matter and potentially some EDCs and PhACs by NF), and membrane
configuration (spiral wound or hollow fiber for UF; spiral wound for NF). With an integrated SAT membrane system, it is envisioned that the only additional treatment required prior to distribution would be disinfection, either with a chemical disinfectant (to provide a residual) or possibly by UV inactivation (to provide an additional barrier).

Another possibility of the integrated SAT-membrane system may be accomplished by membrane treatment followed by SAT, e.g. what is practiced at Orange County Water

Figure 8  SEC-DOC chromatogram of TU-SE feed water compared to permeate by different membranes

Figure 9  SEC-DOC chromatogram of OW2-90 feed water compared to permeate by different membranes

Figure 10  SEC-DOC chromatogram of SIBW-6U feed water compared to permeate by different membranes
District (OCWD), USA, where the final step is really aquifer storage and recovery (ASR). However, we believe a better approach is SAT first for reduction of fouling, and membranes last as a high-integrity second barrier.

Another important issue is whether RO should be selected over NF if lower molecular weight/more polar trace organic compounds (e.g., PhACs and/or EDCs) prove persistent through SAT and are not adequately rejected by NF. In fact, recent observations indicate partial permeation of NDMA (N-nitrosodimethylamine) through RO membranes at OCWD, although very preliminary results suggest NDMA removal during SAT.

**Conclusion**

Soil aquifer treatment (SAT) has been shown to provide significant removal of bulk organic matter (EfOM) from reclaimed wastewater effluent. SAT can be effectively used as a primary barrier in a potable-reuse system. The results have shown that at least 80 per cent of the DOC in wastewater effluent was removed during percolation through the soil. However, some refractory organic compounds still remained in the SAT treated water.

Membrane filtration is an alternative technology for potable reuse. The NF (ESNA) membrane provided higher organic rejection than the UF (GM) membrane due to its characteristics including low molecular weight cutoff (MWCO) and high negatively charged surface. However, the ESNA membrane exhibited higher flux decline due to its properties, including lower MWCO and higher hydrophobicity.

Membrane treatment of SAT treated waters provided a multiple barrier and additional organic matter removal. Additionally, SAT showed a benefit in terms of membrane fouling reduction. According to flux decline tests, it was found that the SAT reclaimed waters showed less flux decline than the corresponding wastewater effluent. The evidence from FTIR spectra and SEC-DOC chromatograms supported the notion that the possible foulants of membranes, e.g. polysaccharide-, protein-like materials, and/or amino sugars, had been reduced during SAT, reflecting the effectiveness of SAT as a pretreatment for membranes. This synergy between SAT and membrane treatment has led to the concept of an integrated SAT-membrane system for potable reuse.

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**References**

