# Treatment of biologically treated landfill leachate with forward osmosis: investigating membrane performance, cleaning protocols and impact of cleaning agents on rejection

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Contents



## Reviewer #1:

In the revised manuscript, the authors revised the manuscript according to all the reviewers comments.I this it is acceptable for publication. There are some points the authors should revise before the formal acceptance.

1. Line 17, the discription of AL-FS mode is wrong.

Action: The description has been corrected in Line 18 Page 1 highlighted in Red colour.

Answer: The revised description is below:

(CTA) membrane for the treatment of landfill leachate (LFL) solution in the active layer facing feed solution and support layer facing the draw solution or AL-FS mode,

2. Line 27, what is the "optimal concentration"?

Action: The optimal concentration has been included. Line 27-29 Page 1.

Response: In the chemical cleaning, an optimal concentration of 3 % hydrogen peroxide was determined for 100% flux recovery of the fouled membrane.

3. LIne 71-73, the sentence is imcomplete.

Answer: The sentence has been completed. Line 70-73 Page 3.

Previous studies studying the impact of membrane orientation on the performance of the FO process suggested that membrane fouling was lower when the dense active layer was facing the feed solution and the support layer faces the draw solution (AL-FS or FO mode) (Tang et al. 2010) compared to when the active layer faces the draw solution and the support layer faces the feed solution or AL-DS mode.

4. Line 139-143, the preparation of H2O2, HCl and NaOH solution should be simply expressed. For example, "the H2O2 solution (30% w/w) was used directly or diluted to 3% w/w, the acid cleaning was performed using HCl solution at pH 4 and the allaline cleaning was conduction using NaOH solution at pH 11."

Answer: The phrases have been modified. Line 137-139 Page 6.

Action: A 30% w/w $H_2O_2$ solution was diluted to 3% w/w and used for the chemical cleaning experiments. Acid cleaning was conducted using aqueous HCl solution at pH 4, and alkaline cleaning was done using aqueous NaOH solution at pH 11.

5. Line 232, the unit of conductivity is mS/cm, not ms/cm.

Answer: The unit has been corrected. Line 226, Page 9.

Action: The conductivity of the raw leachate was 12.10 mS/cm and had a total dissolved solid of 5.5 g/L.

6. Line 307, not Figure 3b and 3c presents, but Figure 3b and 3c present.

Answer: The phrase has been corrected. Line 299, Page 12.

Action: Figure 3b and 3c present the fouling of the membrane in the AL-DS and the AL-FS mode, respectively.

7. The expression should be more concise, some duplicate expression of method should be revised, i.e., Line 465 to 469.

Answer: The expression has been revised and the repetition have been deleted. Line 465-468 Page 18.

Action: Generally, any sequence of acid followed by alkaline or alkaline followed by acid cleaning is an effective approach for the removal of organic and inorganic foulants, which are deposited on the membrane surface.

Reviewer #3:

The authors addressed some of my comments. I still put major revision for two reasons:

1. Still my major concern is representativeness of landfill leachate sample tested in the

study. Landfill leachate is well known for its large variation of characteristics. The findings may not apply to other landfill leachates but the title seems cover all. I understand inclusion of more leachate samples for testing may not be practical.

Answer: Thanks, in general, wastewater characteristics and composition is not consistent over time. The landfill leachate used in all experiments was sampled and analysed for composition. The results should be interpreted that landfill with such composition would be applicable for the FO treatment and cleaning methods would achieve the results provided in the study. This applies for all membrane treatment processes where data show the results based on the membrane used, testing conditions for the feed solution under investigation. The For instance, Textile wastewater in Australia may widely differ in characteristics from a textile wastewater in for instance Middle East. Most studies on forward osmosis landfill leachate have used only one leachate feed solution. A list of few studies are mentioned here for reference (Aftab et al. 2019, Dong et al. 2014, Li et al. 2017, Wu et al. 2018).

2. Still, no statistical analysis of data is a problem to be published in a high impact journal.

Answer: Some statistical data has been incorporated in the supplementary information S.4, Page 4 and Page 5 for flux data. Additional statistics has been incorporated into the XRF analysis and ICP-MS data in Table 1.

**References**:

Aftab, B., Ok, Y.S., Cho, J. and Hur, J. (2019) Targeted removal of organic foulants in landfill leachate in forward osmosis system integrated with biochar/activated carbon treatment. Water Research 160, 217-227.

Dong, Y., Wang, Z., Zhu, C., Wang, Q., Tang, J. and Wu, Z. (2014) A forward osmosis membrane system for the post-treatment of MBR-treated landfill leachate. Journal of Membrane Science 471, 192-200.
Li, J., Niu, A., Lu, C.-J., Zhang, J.-H., Junaid, M., Strauss, P.R., Xiao, P., Wang, X., Ren, Y.-W. and Pei, D.-S. (2017) A novel forward osmosis system in landfill leachate treatment for removing polycyclic aromatic hydrocarbons and for direct fertigation. Chemosphere 168, 112-121.
Tang, C.Y., She, Q., Lay, W.C.L., Wang, R. and Fane, A.G. (2010) Coupled effects of internal concentration polarization and fouling on flux behavior of forward osmosis membranes during humic acid filtration. Journal of Membrane Science 354(1-2), 123-133.
Wu, S., Zou, S., Liang, G., Qian, G. and He, Z. (2018) Enhancing recovery of magnesium as struvite from landfill leachate by pretreatment of calcium with simultaneous reduction of liquid volume via forward osmosis. Science of The Total Environment 610-611, 137-146.

**Treatment of biologically treated landfill leachate with forward osmosis: Investigating membrane performance and cleaning protocols**

List of Tables.

**Table 1:** Main characteristics of LFL wastewater collected from Whyte Gully resource centre, New South Wales

| Parameter | Value | Unit |
|---|---|---|
| Zeta potential | -15.6 | mV |
| Turbidity | 34 | NTU |
| Colour | Light brown | |
| Appearance | Small granules/particulates | |
| pH | 7.82 | - |
| Conductivity | 12.10 | ms/cm |
| TDS | 5550 | mg/L |
| TOC | 149.2±5 | mg/L |
| TC | 204±5 | mg/L |
| IC | 27.6±3 | mg/L |
| Mg | 82.23±5 | mg/L |
| Ca | 66±5 | mg/L |
| K | 429±5 | mg/L |
| Al | 0.08 | mg/L |
| Cr | 0.08 | mg/L |
| Mn | 0.04 | mg/L |
| $Ba^{2+}$ | 0.34 | mg/L |
| Pb | 0.006 | mg/L |
| Si * | 1717±88 | ppm |
| Ag * | 47±7 | ppm |
| Th * | 12±3 | ppm |
| P * | 54±29 | ppm |
| S * | 265±19 | ppm |

* XRF Analysis

Table 2. Flux recovery of fouled membrane after $H_2O_2$ cleaning with different concentrations

| **Membrane orientation** | **$H_2O_2$ Concentration** | **Flux recovery %** |
|---|---|---|
| **AL-FS** | 50ml/L | 75 |
| **AL-FS** | 100ml/L | 102 |
| **AL-DS** | 50ml/L | 62 |
| **AL-DS** | 100ml/L | 91 |

## Supplementary information Tables

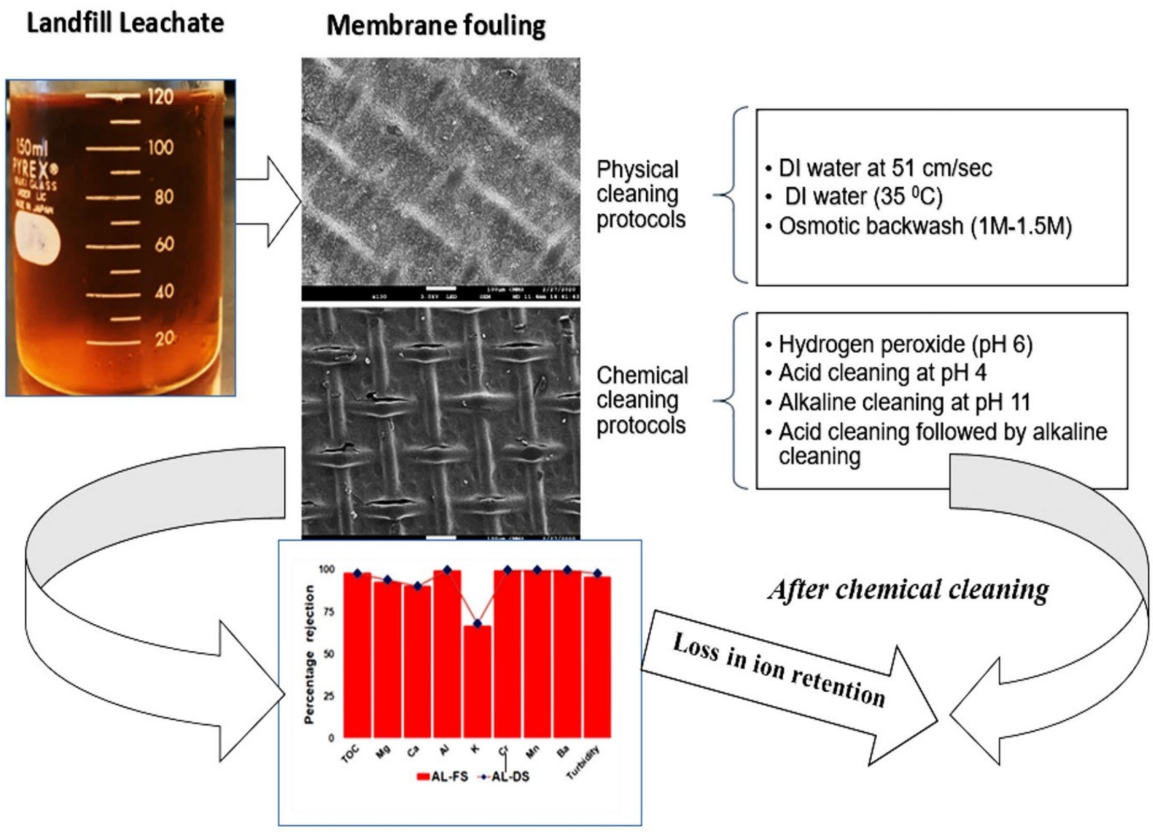
**Table S.1**: Properties of CTA FO membrane

| | |
|---|---|
| Contact angle of the AL | 68.16° |
| Contact angle of the SL | 60.48° |
| Water permeability A | 0.58 L/m$^2$h bar |
| Salt permeability B | 0.25 L/m$^2$.h |
| Salt rejection | 95±1% |
| Chlorine tolerance | 1-2 ppm |
| Maximum operating temperature | 50 °C |

Table S.2. Cummulant results from the Malvern particle analyser

| | |
|---|---|
| Z-average size | 9997 n.m |
| PDI | 0.802 |
| PDI width | 8950 d.nm |

List of Figures




**Graphical abstract.**

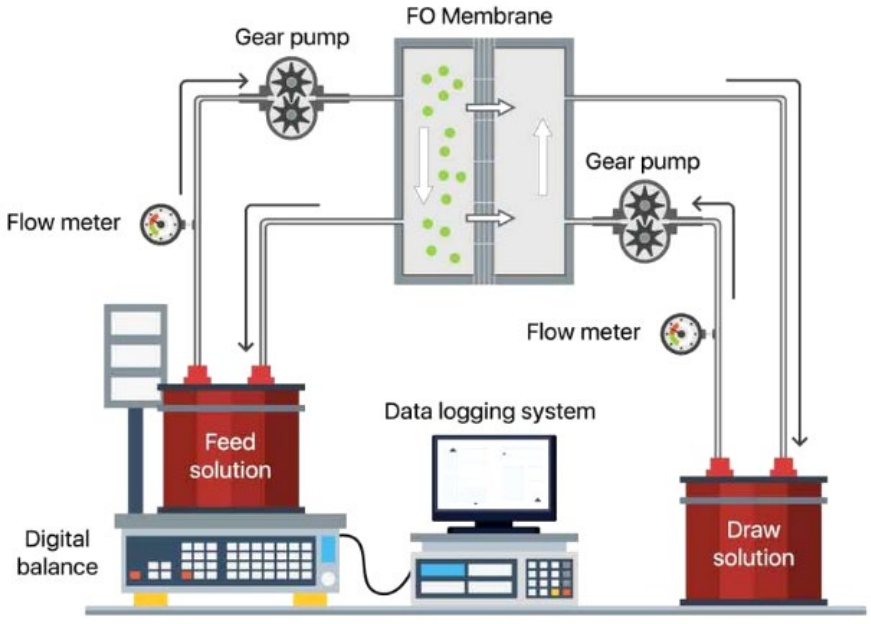


**Figure 1**. Laboratory-scale forward osmosis crossflow filtration unit.

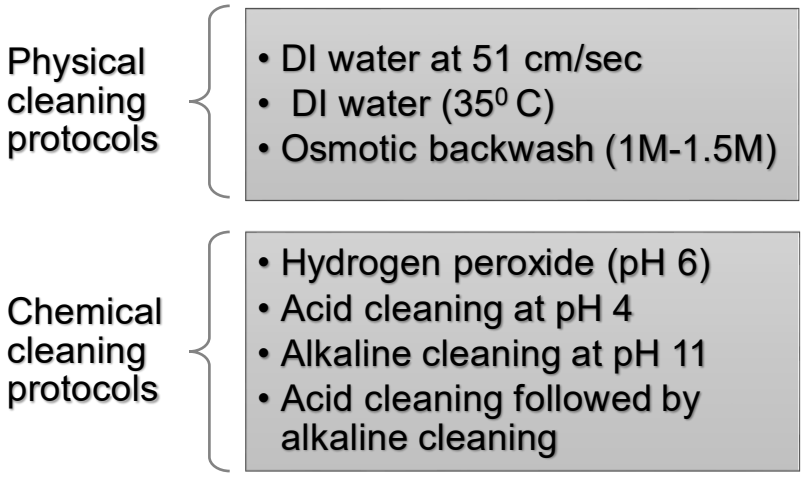



**Figure 2**: Cleaning protocols used in this study in both membrane orientations.

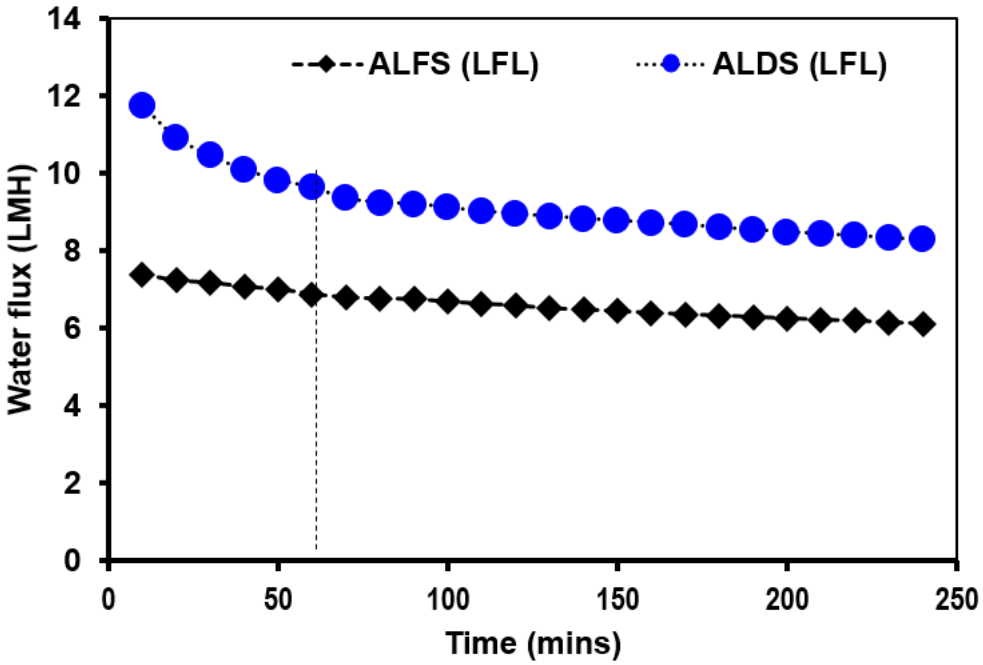



Figure 3a.

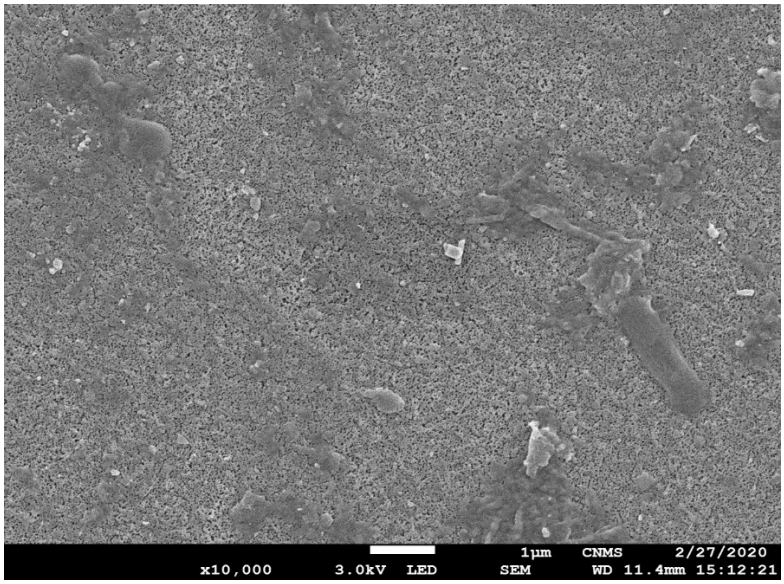



Figure 3b.

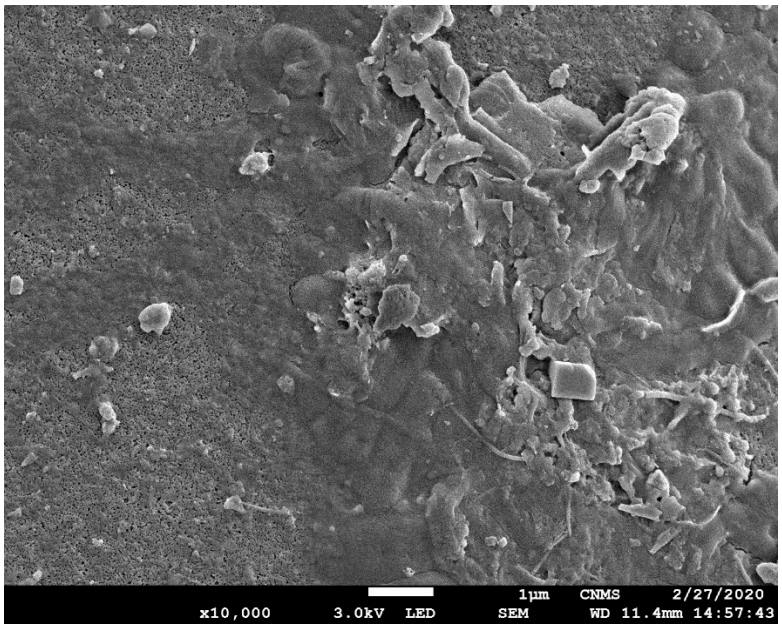



Figure 3c.

**Figure 3: a).**Water flux in the AL-DS mode and the AL-FS with LFL feed, b) fouling in the AL-FS mode on the active layer of the FO membrane, c) fouling in the AL-DS mode on the support layer of the membrane.

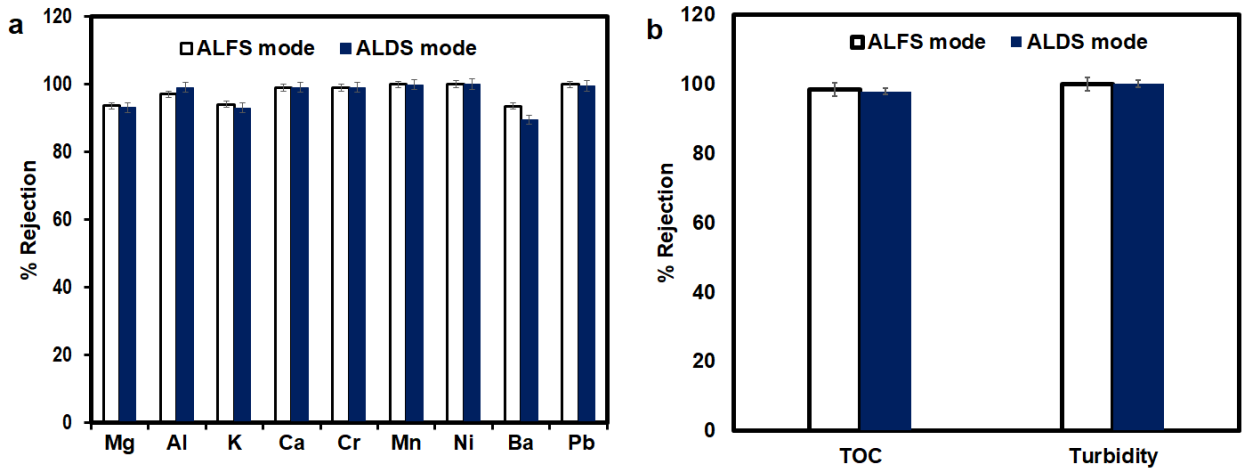
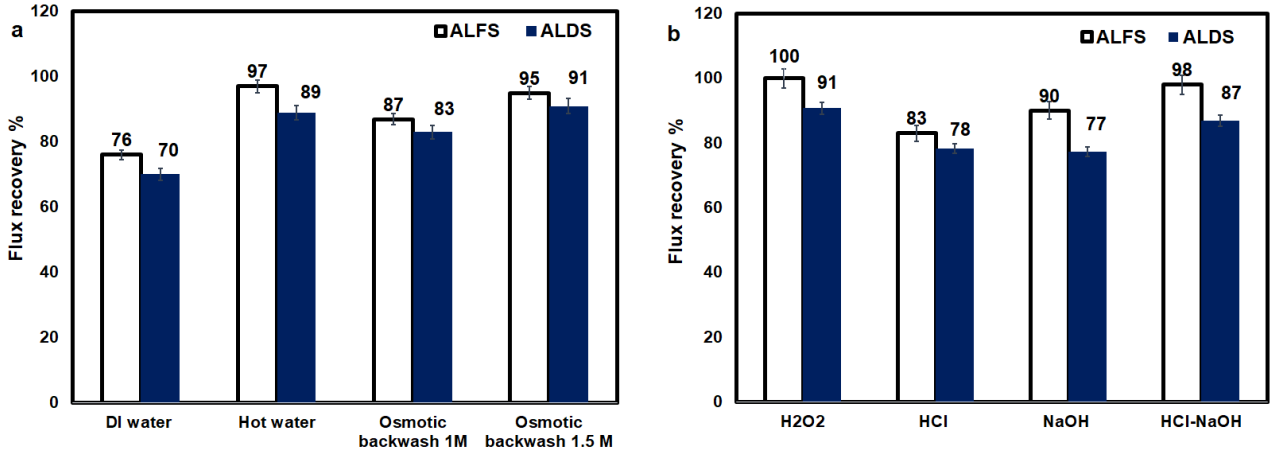



**Figure 4**: a). Rejection of CTA FO membrane in AL-FS and AL-DS mode analysed by ICP-MS, b). Rejection of CTA FO membrane for TOC and turbidity analysed by TOC analyser and Turbidity meter. * The amount of Ni was not detectable in the permeate solution and therefore almost 100% rejection for Ni for the FO membrane.



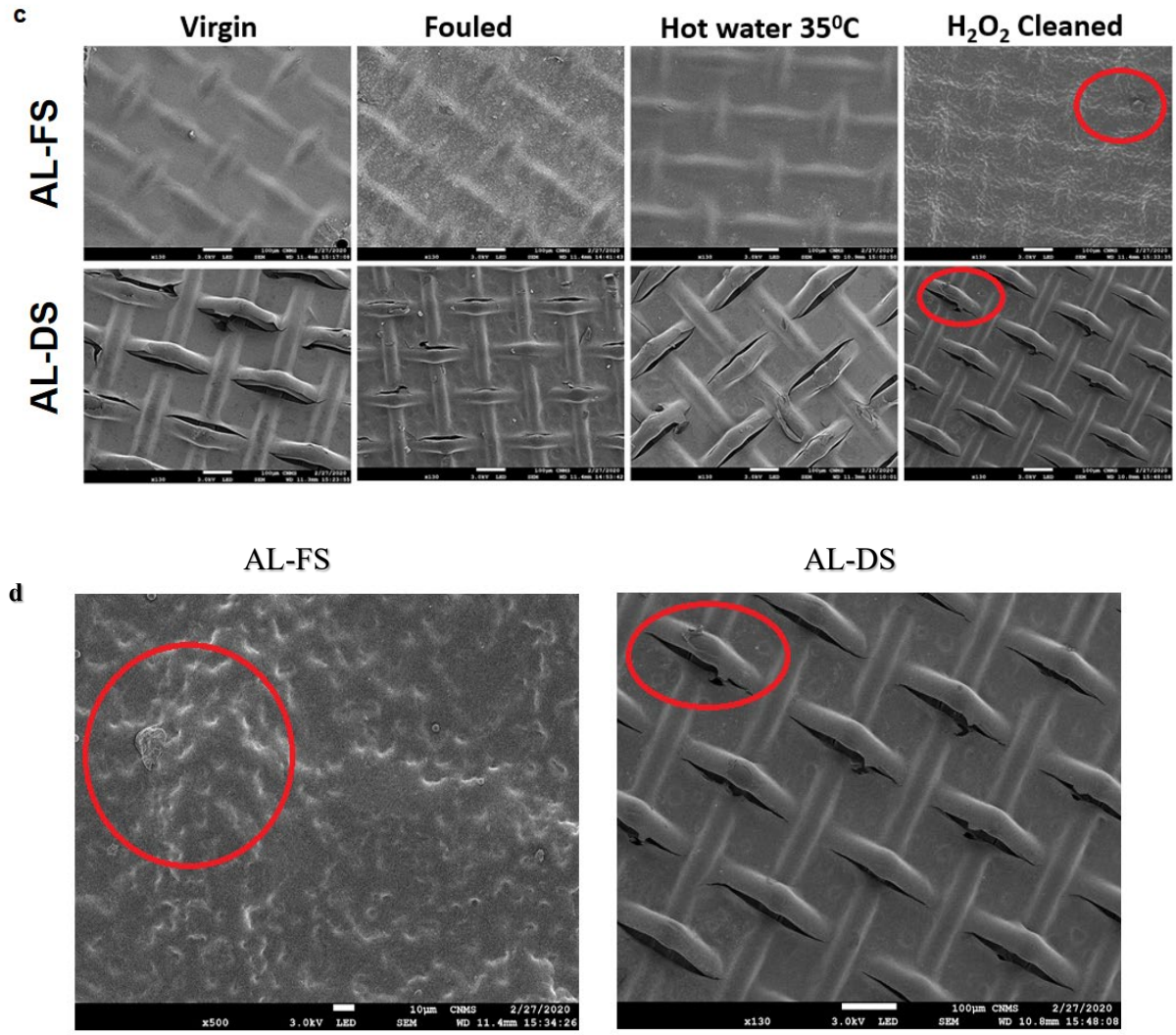



**Figure 5**) Comparison of physical and chemical cleaning protocols for CTA FO membrane. a). Physical cleaning protocols (DI water at 51cm.sec$^{-1}$, Hot water 35$^{o}$C, osmotic backwashing 1M, and 1.5M) in AL-FS and AL-DS mode. b). Chemical cleaning protocols ( 100mL/L $H_2O_2$, 0.05M HCl at Ph 4, 0.05M NaOH at pH 11, and sequential cleaning, which was 0.05M HCl at pH 4 followed by 0.05M NaOH at pH 11) in AL-FS mode and AL-DS. c). FE-SEM images of the virgin membrane, fouled membrane, hot water 35$^{o}$C cleaned, and $H_2O_2$ cleaned membrane, d). FE-SEM of membrane damaged by $H_20_2$. The red circle in $H_2O_2$ cleaned membrane illustrates a slight peeling of the membrane layer due to chemical cleaning. d) Damage on the active layer by hydrogen peroxide cleaning, a). On the active layer in the AL-FS mode. b). Slight peeling of the support layer in the AL-DS mode.

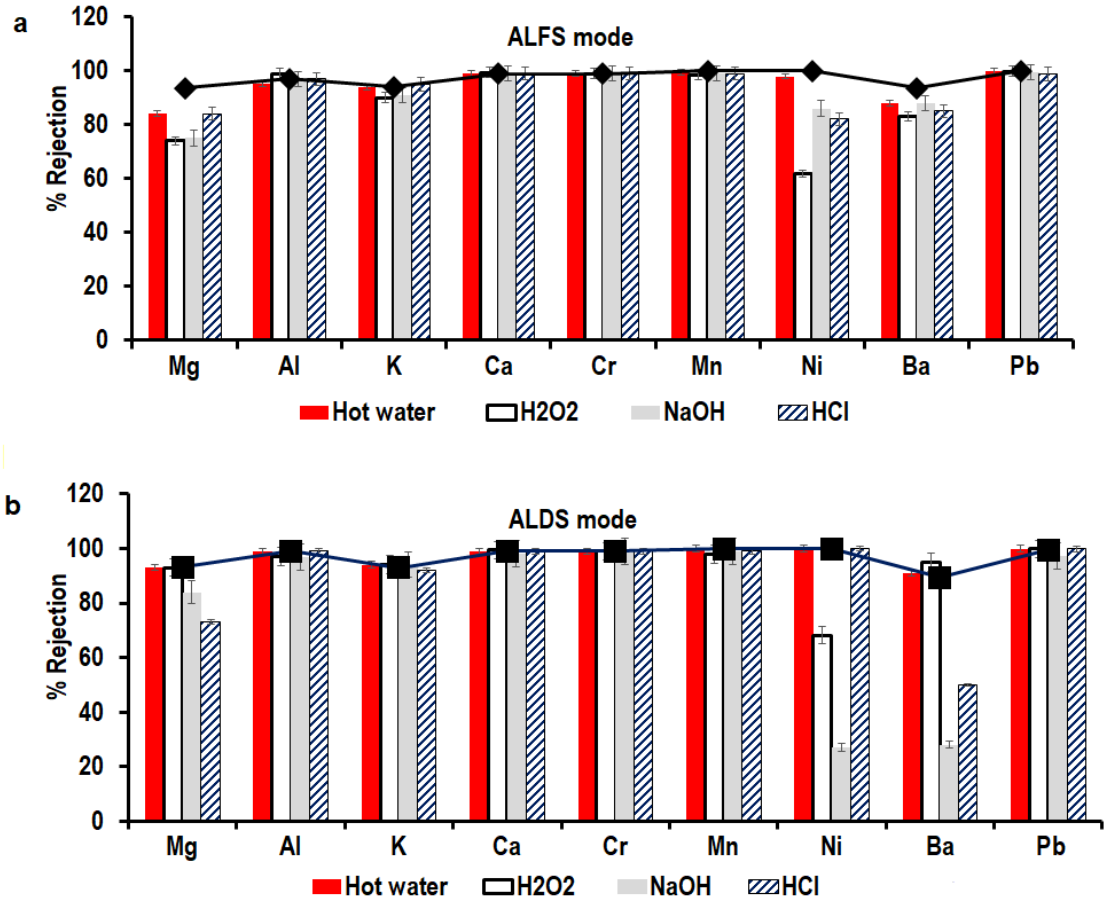



**Figure 6:** a). Impact of physical and chemical cleaning protocols on pollutants rejection in the AL-FS. b). Impact of physical and chemical cleaning protocols on pollutant rejection in the AL-DS. Physical cleaning protocols included Hot water cleaning at 35°C. Chemical cleaning protocols were 100ml/L $H_2O_2$ , 0.05M HCl cleaning at pH 4, and in 0.05M NaOH at pH 11. For Ni rejection, no column or zero rejection represents Ni was not detected in the samples.

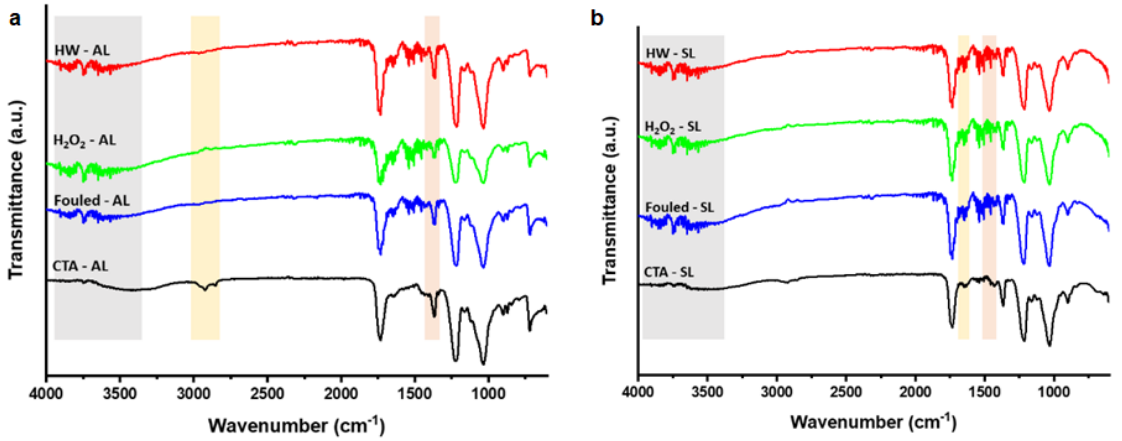



**Figure 7**: FT-IR spectra of the virgin, fouled, and cleaned membrane a) Active layer of the FO membrane in the AL-FS mode b) Support layer of the FO membrane in the AL-DS mode. Hot water cleaning (HW).

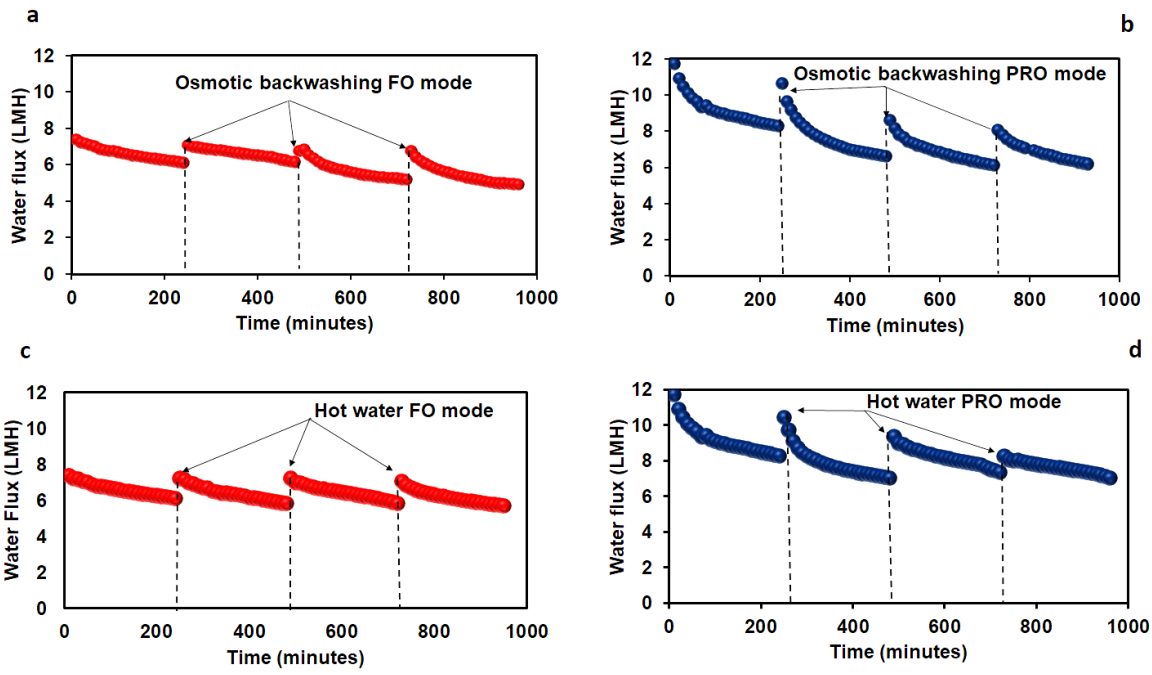



**Figure 8:** Physical cleaning protocols employed over four cycles of FO operation. a) Osmotic backwashing with 1.5M NaCl in the AL-FS mode, b) Osmotic backwashing with 1.5M NaCl in the AL-DS mode. c) Hot water 35 ºC cleaning in the AL-FS mode, d) Hot water 35 ºC cleaning in the AL-DS mode.

## Supplementary information Figures

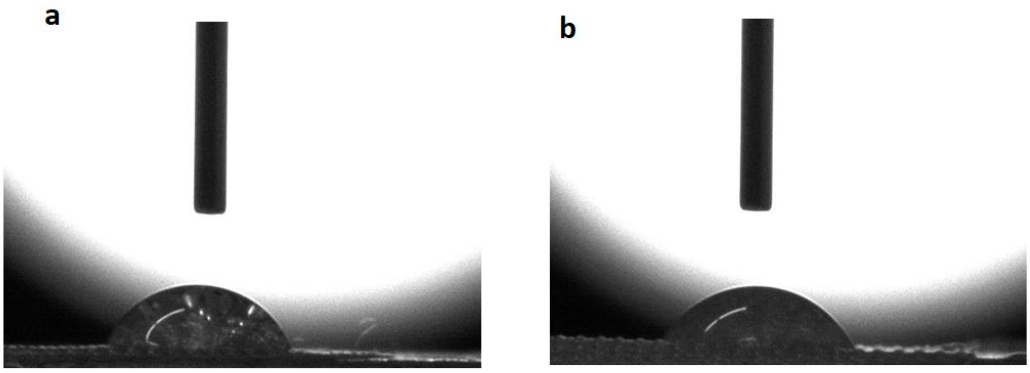



Figure S.1. a). Contact angle of the active layer. b). Contact angle of the support layer.


**Figure S.2**. Colour of the landfill leachate feed, indicating the presence of humic substances.

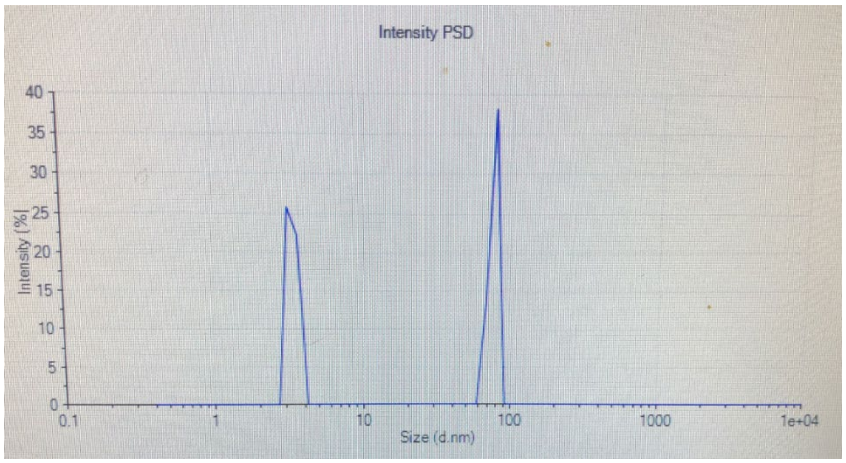



Figure S.3. Size distribution of particles in the LFL wastewater

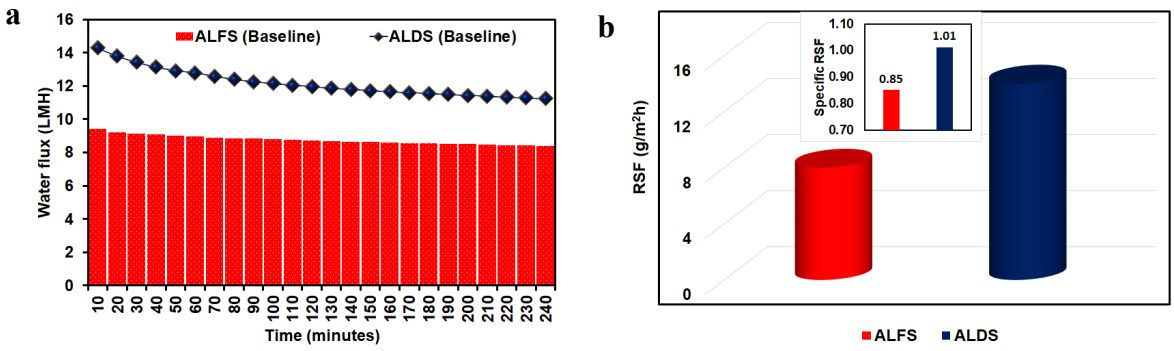



**Figure S.4** a) Water flux of CTA membrane in FO and PRO mode with DI water feed and 0.6M draw solution. **S.3**b) Reverse solute flux of CTA membrane in FO and PRO mode with DI water feed solution and 0.6M draw solution.