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Article

Permeability is the Critical Factor Governing the Life Cycle Environmental Performance of Drinking Water Treatment Using Living Filtration Membranes

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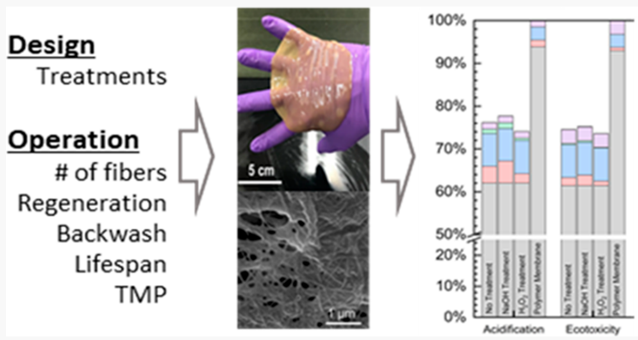

ABSTRACT: Living Filtration Membranes (LFMs) are a water filtration technology that was recently developed in the lab (Technology Readiness Level 4). LFMs have shown filtration performance comparable with that of ultrafiltration, far better fouling resistance than conventional polymer membranes, and good healing capabilities. These properties give LFMs promise to address two significant issues in conventional membrane filtration: fouling and membrane damage. To integrate environmental considerations into future technology development (i.e., Ecodesign), this study assesses the life cycle environmental performance of drinking water treatment using LFMs under likely design and operation conditions. It also quantitatively ranks the engineering design and operation factors governing the further optimization of LFM environmental performance using a global sensitivity analysis. The results suggest that LFMs' superior fouling resistance will reduce the life cycle environmental impacts of ultrafiltration by 25% compared to those of a conventional polymer membrane in most impact categories (e.g., acidification, global warming potential, and carcinogenics). The only exception is the eutrophication impact, where the need for growth medium and membrane regeneration offsets the benefits of LFMs' fouling resistance. Permeability is the most important factor that should be prioritized in future R&D to further improve the life cycle environmental performance of LFMs. A 1% improvement in the permeability will lead to a ∼0.7% improvement in LFMs' environmental performance in all the impact categories, whereas the same change in the other parameters investigated (e.g., LFM lifespan and regeneration frequency) typically only leads to a <0.2% improvement.

■ INTRODUCTION

Recently, Living Filtration Membranes (LFMs) were successfully developed as a water filtration technology for the first time using native microorganisms of a kombucha symbiotic culture of bacteria and yeast.[1] Composed of a bacterial cellulose (BC) network with embedded microorganisms, LFMs have physical properties highly suitable for water filtration (e.g., high tensile strength and hydrophilicity[2,3]). In bench-scale dead-end filtration tests, LFMs have permeability and size cutoff values similar to commercial ultrafiltration membranes (i.e., 135 ± 25 L $m^{-2}h^{-1}bar^{-1}$ and a 90% rejection of 30 nm).[4]

Most notably, LFMs have demonstrated high fouling resistance. When treating the influent used by the Basin Creek Drinking Water Treatment Plant (Butte, MT, USA), the flux loss with LFMs was only 50% after 7 h of operation, considerably lower than the >90% flux loss experienced by a commercial polymer membrane (Millipore VMWP02500, Massachusetts, USA) after the same time (unpublished results attached for review only). This suggests that LFMs can potentially address the greatest challenge in membrane filtration: membrane fouling. Fouling greatly reduces the efficiency of membrane filtration and can occur even under harsh conditions.[5,6] Despite the high attention given to membrane fouling, there have been limited breakthroughs in its mitigation.[7−10]

In addition to fouling resistance, LFMs have also demonstrated great healing capabilities. Even after severe damages (e.g., 3 mm holes), LFMs achieved 75−80% recovery of flux in a period of 4−17 days simply by being placed in growth solutions at 25 °C.[1,4,11] The healing capabilities of LFMs have the potential of addressing another common operational challenge in membrane filtration: fiber damage.[12,13] While the frequency of fiber damage may be moderate,[14] it is an operational nuisance, and a number of common operating conditions (e.g., chemical cleaning) can increase the frequency of its occurrence.[15]

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LFMs have created a new category of water filtration technologies, as they are a biodegradable material that may be fabricated into virtually any shape from μm to mm dimensions,[16] and may be structurally and chemically modified to impregnate antifouling[17,18] and/or diffusion properties.[4,19] A multitude of subsequent studies can be developed upon this proof of concept, e.g., permeability modifications,[11] functional modifications through incorporation of other molecules,[20−22] and optimization of the engineering design and operation conditions.[23]

To integrate environmental considerations into future technology development (i.e., Ecodesign),[24,25] herein, we conduct a preliminary life cycle assessment of LFMs as a drinking water filtration technology. Since LFMs are currently in the lab development stage, we extrapolate its potential future full-scale environmental performance based on a combination of lab data (e.g., membrane characteristics, such as permeability and regeneration frequency) and real-world operational data (e.g., backwash frequency, and chemical and energy consumption) from the Basin Creek Drinking Water Treatment Plant (Butte, MT, USA). We then identify high-priority design parameters using a global sensitivity analysis over a comprehensive design space informed by the conditions accomplished in the lab and the full-scale plant in Butte, MT.

■ METHODS AND DATA

Overview of LFMs. A detailed, technical description of LFMs is available elsewhere and in the SI.[4] Briefly, LFMs currently are a lab-scale water filtration technology (Technology Readiness Level 4). They have successfully treated deionized water at the 100 L/day scale and achieved filtration performance comparable to that of ultrafiltration with a size cutoff of 30 nm.[11]

Goal of the Life Cycle Assessment. The goal of this study is to estimate the life cycle environmental impacts of LFMs in a full-scale drinking water operation, and to the extent possible, compare them with those of conventional full-scale membrane filtration technologies to assess whether LFMs can reduce the environmental impacts associated with drinking water treatment in their current state. A functional unit (FU) of 1 MGD (4645m^3/day) treated water is thus chosen. Two things should be clarified. First, again, since LFMs are still in the early stages of development, this LCA is indicative of the current technology readiness level (TRL 4) rather than a more mature level that would be expected in commercial production. Second, this LCA assessment has not been certified by an independent LCA analyst and is not intended to be used for any commercial or marketing purposes.

Scope of the Life Cycle Assessment. The scope of this LCA study is from raw material extraction through to end-of-life waste disposal (Figure 1). Included in the system boundary are growth medium production, LFM production, LFM module assembly, LFM module operation, and end-of-life disposal (Figure 1). Key upstream and downstream processes, such as electricity production and end-of-life disposal of LFMs, are included. Construction of the water treatment facilities (e.g., concrete and pipes) and transportation are excluded. The omission of construction and transportation likely has insignificant impacts, since all previous studies reported that the life cycle impacts of conventional membrane filtration plants are dominated by the operation stage.[26−28] The LFM modules (plant) are assumed to operate 20 h/day for 300 days/year, with a lifespan of 20 years.

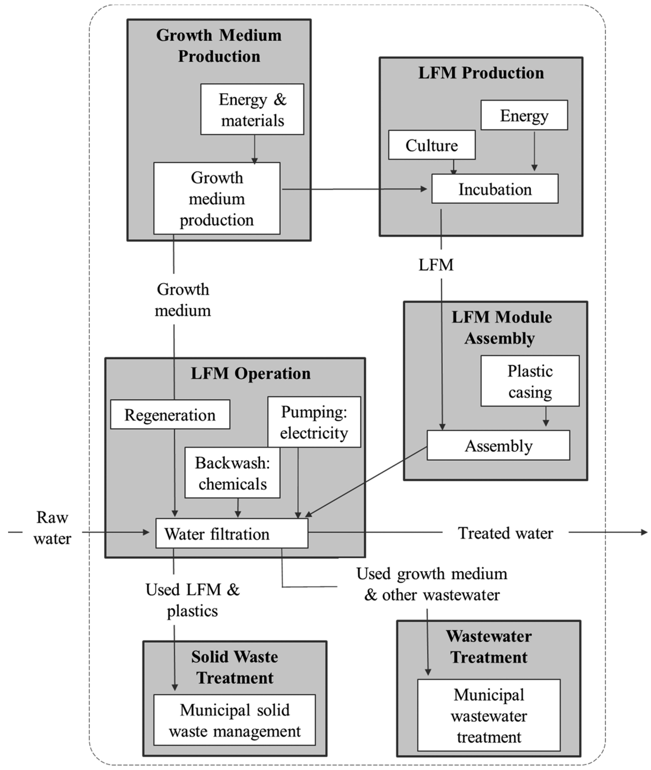

Figure 1. Overview of LFM-based water filtration. Dotted box is the system boundary. The treated water produced by LFMs does not require modifications to subsequent drinking water treatment processes (e.g., disinfection) based on current knowledge.

This LCA study compares LFMs with a mixed cellulose esters polymer membrane (Millipore VMWP02500, Massachusetts, USA, 0.05 μm pore size), which is picked based on similarities with LFMs in terms of filtration performance (i.e., cutoff and permeability). Identical FU, plant life span, and operation time are used. The same life cycle stages are included. Growth medium production, LFM regeneration and LFM replacement are excluded, as they are not needed for polymer membranes.

Life Cycle Inventory. The inventory data used are a combination of lab data (Table S1), real-world operation data from a full-scale membrane filtration plant (Table S2), and Ecoinvent v3.4 (Table S3). The lab data (foreground data) primarily include the relationships between flux and transmembrane pressure (TMP) under different treatments, the development of fouling and its impact on flux over time, the material and energy consumption in the growth medium production, and the frequency and material and energy consumption in LFM regeneration. The full-scale operation data (foreground data) are based on the Basin Creek Water Treatment Plant in Butte, MT, which include the frequency of backwash and the material and energy consumption in backwash. Ecoinvent is used as the background database, which include data such as the impacts of wastewater treatment and electricity supply (Table S3).

Life Cycle Impact Assessment. The U.S. EPA's Tool for the Reduction and Assessment of Chemicals and Other Environmental Impacts (TRACI 2.1 version 1.02) was used to assess the impact of LFM filtration in OpenLCA (v1.8). All of its midpoint impact category indicators are reported. The results are grouped into module production, operation—

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Table 1. Design Parameters and the Ranges of Values Studied in the Sensitivity Analysis[a]

			Flux-TMP/time relationships	
LFM Property	*Permeability*		$Flux_0 = A \times TMP$	
	No treatment			A = 119.9 L/(m^2 h bar)
	0.3% H_2O_2			A = 217.7 L/(m^2 h bar)
	0.5 M NaOH			A = 89.4 L/(m^2 h bar)
	Fouling Resistance		$Flux_t = Flux_0 \times [\exp(a \times t)]$	
	LFMs			a = −0.265 h^{-1}
	Polymer membrane			a = −1.233 h^{-1}
		Baseline values	**Range simulated in global sensitivity analysis**	**Accomplished in lab or real-world operation**
Design/Operation Parameters	*Module Assembly*			
	Number of fibers per module	800	(400, 1800)	NA
	Maintenance			
	LFM regeneration frequency (times/year)	40	(20, 90)	(20, 60)
	Backwash frequency (flux loss)	15%	(10%, 45%)	10−15%
	Durability			
	LFM fiber lifespan (year)	0.8	(0.4,1.8)	NA
	Operation			
	TMP (bar)	4	(0.7, 5.6)	(0.7, 3.1)

[a]The flux-TMP relationship is based on the work in Song 1998.[29] The fouling resistance is based on lab results. A: membrane-specific constant; TMP: trans-membrane pressure; $Flux_t$: flux at time t; $Flux_0$: flux at time 0; t: time, a: constant; NA: not available.

backwash, operation−electricity, operation−regeneration, end-of-life (EoL)−wastewater, and EoL−others. Module production includes the material and energy consumption associated with producing LFM modules and fibers (e.g., plastics, growth medium etc.). "Operation−backwash" includes the energy and chemicals needed for all three types of backwash. "Operation−electricity" is the electricity consumption during operation (i.e., primarily pumping). "Operation−regeneration" includes the materials and energy needed for LFM regeneration (e.g., the growth medium needed). "EoL−wastewater" is the treatment of the wastewater generated during operation, which includes the growth medium used in regeneration and 1% of FU (i.e., 99% recovery assumed). "EoL−others" include the disposal of EoL plastics and LFMs.

Design Parameters. Two types of parameters are selected to understand the potential of improving the life cycle environmental impacts of LFMs through engineering design and operation: LFM property (i.e., permeability and fouling resistance) and design/operation parameters (Table 1). These parameters are directly related to the filtration performance and the material/energy consumption of LFMs, according to lab results and/or theoretical understandings of the mechanisms.

Permeability is an LFM property that determines the flux of LFMs under a certain TMP. It was shown that LFMs have the capability of undergoing chemical treatments to achieve different permeability and selectivity.[11] This capability has direct engineering and environmental implications. Higher permeability will reduce the energy consumption of LFM filtration (and thereby likely reducing the environmental impacts of LFM filtration), while higher selectivity will allow LFMs to adapt to changing source water qualities. Herein, we explore the environmental impacts of three LFM treatments (i.e., two chemical treatments and a no treatment baseline). The flux-TMP relationship for each treatment is extrapolated from lab results and shown in Table 1.[11]

Fouling resistance is an LFM property that characterizes how the flux decreases as fouling develops. In preliminary lab results, LFMs demonstrated a considerable advantage over conventional membranes. With the real influent used by the Basin Creek Drinking Water Treatment Plant in Butte, MT, the flux loss was 50% with LFMs after 7 h of operation, as opposed to a >90% flux loss with a commercial polymer membrane after the same operation time (unpublished results attached for review only).[11] The fouling resistance of LFMs is modeled using a time-dependent equation based on lab results (a = −0.265 and −1.233 h^{-1} for LFMs and polymer membrane, respectively, Table 1). The LFMs are assumed to operate in the constant-flux mode, which has advantages over the constant-TMP mode and is popular in industrial applications.[30,31]

Module assembly explores the sensitivity of environmental performance to the number of LFM fibers assembled into each module. A typical hollow-fiber design often used in commercial ultrafiltration is followed (length 2m, diameter 0.2m). Each LFM fiber is assumed to be 2 m in length and 4 mm in diameter, with a wall thickness of 1.5 mm (LFM fibers are currently produced as 1.5 mm-thick flat sheets in the lab). With this design, the theoretical maximum number of LFM fibers per module is ∼2500. The number of fibers per module simulated in this analysis is (400, 1800) (Table 1). A conservative range is chosen given the lack of demonstrated success. The impact of increasing this parameter can be inferred from the global sensitivity analysis.

Maintenance first includes the periodic regeneration of LFMs to maintain their structural robustness, which can be done by filling each module with growth medium at designated frequencies during the down time. Given the size of the modules, 63 L of growth medium is needed for each regeneration. The range simulated in this study is (20, 90) times per year (Table 1). The most feasible regeneration frequency needs to be further investigated in future research and development.

Also included in maintenance is backwash, which includes three types: daily backwash with reverse water flows, weekly chemical cleaning with NaClO, and semiannual cleaning with

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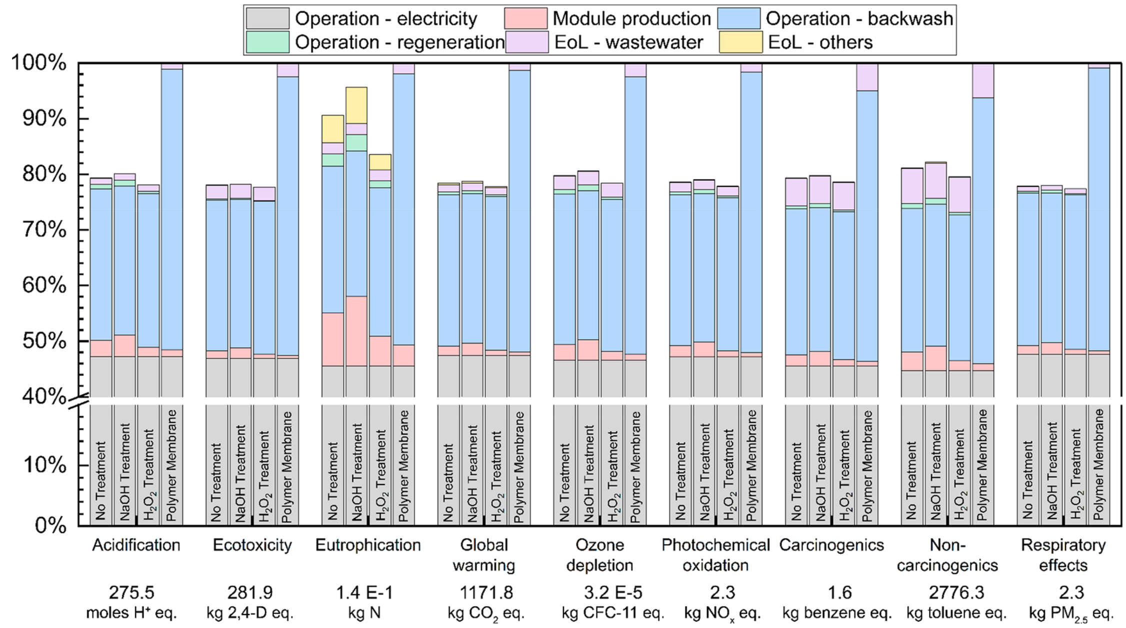

Figure 2. Relative contributions to each midpoint impact category by process and chemical treatments in LFMs and polymer membrane. The un-normalized 100% impacts are impacts of polymer membrane per FU and the numbers are shown below the *x*-axis.

NaClO and citric acid. Daily backwash is initiated once the flux decreases to a certain fraction of the initial flux (e.g., backwash every time the flux decreases to 85% of the initial flux). The values used are specified in Table 1.

Weekly chemical cleaning and semiannual cleaning are modeled using real-world operation data from the Basin Creek Water Treatment Plant in Butte, MT (which uses ceramic membranes), due to the lack of LFM-specific lab data. It is possible that LFMs do not require or could not withstand the same chemical treatments as polymer or ceramic membranes. The real-world operation data are followed in this study as it is the best available data. Further LMF-specific research is needed in the future.

Durability characterizes the frequency of LFM fiber replacement, which is dependent on the lifespan of LFMs, the conditions, and the water treated. Lab data on the range of LFM lifespan is not yet available. An arbitrary range of 0.4−1.8 years is explored.

Operation involves the application of a TMP to generate treated water. The range of TMP accomplished in the lab is (0.7, 3.1) bar for LFMs. The range simulated in this study is (0.7, 5.6) bar, which covers the typical pressure applied in the full-scale plant in Butte, MT (4−5 bar). The flux under each TMP and treatment is modeled using the flux-TMP relationships specified in Table 1.

The parameters used in the "baseline" case are 800 fibers per module, regeneration frequency 40/year, replacement frequency 0.8/year, TMP 4 bar, and backwash 4 times per day (5 h/run), which are based on what is typically accomplished in the lab (e.g., regeneration frequency) and full-scale operation data (e.g., TMP and backwash frequency). The commercial polymer membrane used as a reference is based on the design and operation conditions typically accomplished in commercially available filtration modules and full-scale plants, namely 2000 fibers per module, no regeneration needed, replacement frequency 0.1/year, TMP 4 bar, and reverse water flow backwash when there is a 15% loss of flux.

Sensitivity Analysis. A global sensitivity analysis is conducted by combining Latin−Hypercube (LH) sampling[32] and One-factor-At-a-Time (OAT) approach[33] as outlined in a previous study.[34] The LH-OAT analysis is done for each chemical treatment (Table 1). Within each chemical treatment, $M(e_1, ..., e_P)$ is one of the midpoint impact categories in TRACI (e.g., acidification) that depends on P parameters (here, P = 5, i.e., the five design/operation parameters detailed in Table 1). Each parameter e_i is assumed to be uniformly distributed on an interval $[a_i, b_i]$ (shown in Table 1) and divided into N (here, N = 7) strata with a probability of occurrence equal to 1/N in the LH sampling method. Thus, the total parameter space is divided into N^P (here, $7^5 = 16807$) LH cubes. In each LH cube, one random sample of the parameters $(e_1,\cdots, e_P)$ is generated, and the partial effect of parameter e_i on the impact factor M is calculated by OAT approach (eq 1)

$$S_i = \frac{1}{f_i}\cdot\frac{M(e_1, \cdots, e_i*(1+f_i), \cdots, e_P) - M(e_1, \cdots, e_i, \cdots, e_P)}{[M(e_1, \cdots, e_i*(1+f_i), \cdots, e_P) + M(e_1, \cdots, e_i, \cdots, e_P)]/2} \quad (1)$$

where S_i is the sensitivity and f_i is the fraction by which the parameter e_i is changed (a predefined constant with value 10^{-5} for all (i). P + 1 evaluations of M are then conducted at each LH point, and the total simulation requires $(P + 1)*N^P$ evaluations of M.

RESULTS AND DISCUSSION

Comparison of LFMs and the Commercial Polymer Membrane Used in This Study. In the "baseline" projection, LFMs outperform the polymer membrane by 20−25% in all but one of the impact categories (Figure 2). The advantage is primarily attributable to LFM's superior fouling resistance and the resultant decrease in the electricity consumption during operation. The exception is eutrophication impacts, for which LFMs only show a 3−18% improvement over the polymer membrane (Figure 2), mainly because the electricity savings are offset by the increased impacts from the production of growth medium and LFM fibers during the

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Table 2. Comparison with the Life Cycle Global Warming Potential Impacts of LFMs (Baseline Projection) and Membrane Filtration Technologies Reported in Previous Studies[a]

Reference (technology readiness)	GWP per m^3 treated (kg CO_2 eq/m^3)	LCIA model	Water treated	Note
This study (lab scale)	0.20	TRACI 2.1	DW	LFMs, excluding construction and pretreatment (coagulation, prefiltration etc.)
This study (lab scale)	0.25	TRACI 2.1	DW	VWMP6 polymer membrane, excluding construction and pretreatment (coagulation, prefiltration etc.)
Ribera et al. 2014[26] (full scale)	0.13−0.15	Recipe Midpoint (H)	DW	Nanofiltration
Bonton et al. 2012[28] (full scale)	0.04	Impact 2002+	DW	Nanofiltration
Tangsubkul et al. 2006[35] (full scale)	0.03−0.30	Unspecified	WW	Microfiltration, including different operating conditions such as TMP and flux
Carre et al. 2017[27] (full scale)	0.25	CML	WW	Ultrafiltration
	0.41			Ultrafiltration + UVB
	0.42			Microfiltration + UVD
Ortiz et al. 2007[36] (full scale)	0.02−0.5	CML Eco-indicator 99 Ecopoints 97	WW	Ultrafiltration, estimated based on the difference between two processes: activated sludge, and activated sludge + ultrafiltration
Godskesen et al. 2013[37] (full scale)	1.18	EDIP 1997	DW	Ultrafiltration, including pretreatment, desalination, and UV

[a]DW: drinking water. WW: wastewater.

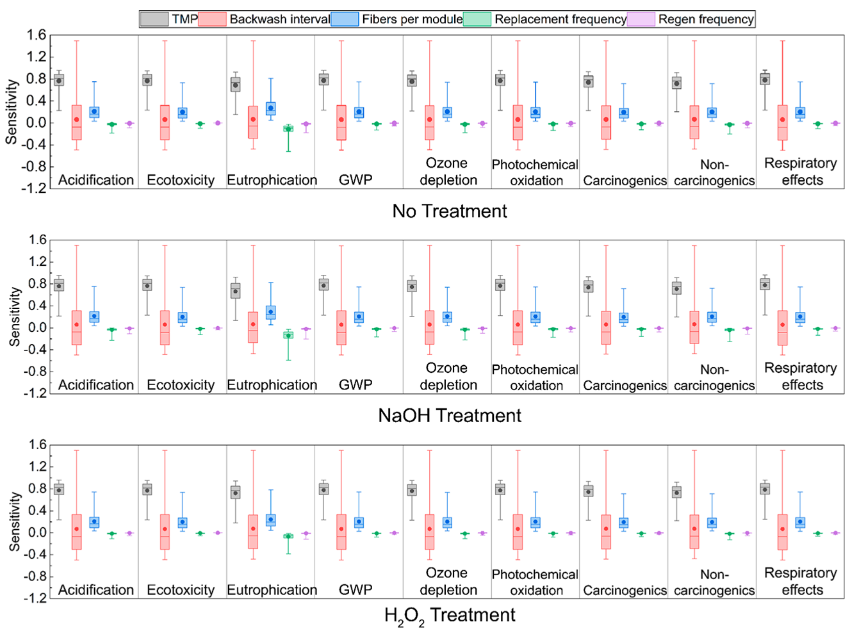

Figure 3. Sensitivity of LFMs' environmental performance to each parameter and treatment.

initial module production and the LFM regeneration in operation. Different LFM treatments show limited impact on the life cycle impacts of LFMs in all of the impact categories (i.e., < 3%), except for eutrophication, where different LFM treatments can lead to a 20% difference (Figure 2).

In terms of relative contributions by process, LFMs and polymer membrane are similar in that the electricity consumption during operation and backwash is the largest contributor to most impact categories (Figure 2). For LFMs, the operation and backwash typically account for >85% of the total impact in all categories; for polymer membranes, the percentage contributions of the two processes are over 95% (Figure 2). The exception again is the eutrophication impact, in which the other processes (such as regeneration and EoL) can account for up to 25% of the total impact (Figure 2).

Comparison of LFMs with Conventional Membrane Filtration in Previous Studies. The life cycle impacts of LFM filtration are also compared with previous studies on polymer membranes (Table 2). Despite the differences in the technology (e.g., ultrafiltration vs nanofiltration), the unit processes included (e.g., with or without construction and pretreatment), and the LCIA method, a general observation is that the life cycle GWP impacts of LFMs and the reference polymer membrane used in this study do not significantly exceed that of previous studies on the basis of per m^3 water treated (Table 2). A comprehensive validation should be attempted when more information becomes available.

Global Sensitivity Analysis. The global sensitivity analysis assesses the importance of each parameter in terms of further improving LFM environmental performance by quantitatively ranking the sensitivity of each parameter over a comprehensive design space. The result suggests that TMP (permeability) is the most critical design/operation parameter to further reduce the life cycle environmental impacts of LFMs.

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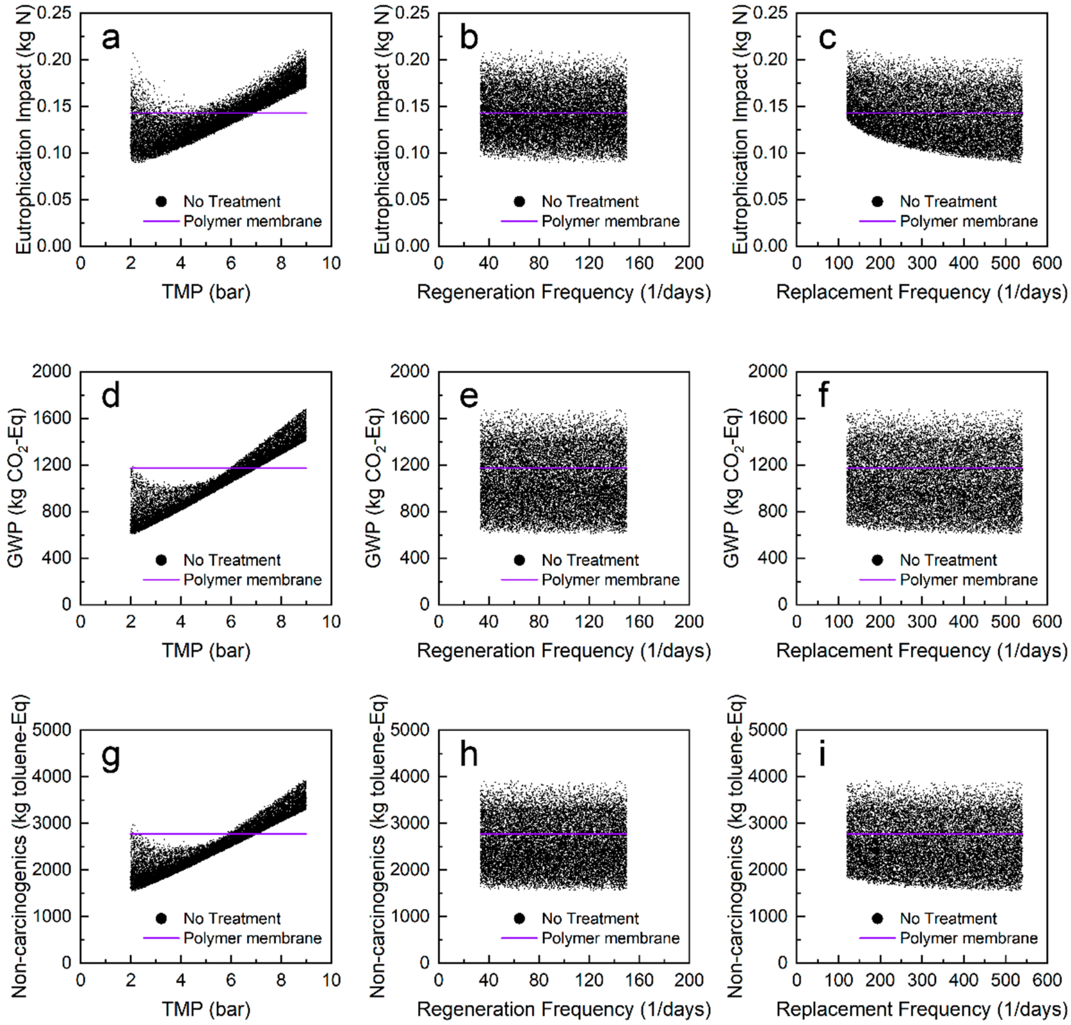

Figure 4. Sensitivity of GWP, eutrophication, and noncarcinogenics to TMP, regeneration frequency, and replacement frequency with untreated LFMs. Each black dot is the result of one simulation. The purple line in all the figures is the respective impact of polymer membranes under 2000 fibers per module, no regeneration needed, replacement frequency 0.1 times/year, TMP 4 bar, and backwash at 15% flux loss.

Changing the TMP by 1% typically results in a 0.6−0.9% change in the life cycle impacts of LFMs, which is statistically significantly higher ($p < 0.05$) than the sensitivities of the other four parameters in most of the impact categories (Figure 3). For example, with untreated LFMs, reducing the TMP by ∼0.04 bar will reduce the GWP impact by ∼7.0 kg CO_2 eq/FU, whereas changing the LFM replacement frequency by 1% only changes the GWP impact by ∼0.5 kg CO_2 eq/FU (Figure 3).

In a comparison of the three treatments, H_2O_2 treatment dampens the sensitivity of LFMs' life cycle impacts to TMP as it increases the permeability of LFMs, whereas NaOH treatment enhances the sensitivity as it decreases the permeability of LFMs (Figure 3). The changes in sensitivities due to treatments, however, are typically insignificant (<2%). For example, the sensitivity of GWP to TMP is 76.0%, 77.4%, and 77.8% for untreated, NaOH-treated, and H_2O_2-treated LFMs, respectively (Figure 3).

To reduce the eutrophication impacts of LFMs, which is the worst performing category of LFMs relative to that of the polymer membrane (Figure 2), an increase in the permeability is still the most effective measure (Figure 3). A 1% improvement in permeability can reduce the eutrophication impacts of LFMs by ∼0.6% across all treatment types (Figure 3). This is aligned with the results that electricity remains the biggest contributor to LFMs' eutrophication impacts, despite the increased relative importance of LFM fiber production, regeneration, and disposal (Figure 2). To reduce the impacts of those processes, new engineering ideas are needed, because simply reducing the frequency of LFM fiber regeneration and replacement has little impact on the overall eutrophication impact (sensitivities ∼0 in Figure 3).

One-Factor-at-a-Time Sensitivity Analysis. All 16807 simulation results are visualized to assess whether the sensitivity of each parameter on final impacts is range-dependent. Figure 4 confirms that the sensitivity of life cycle impacts to the TMP, regeneration frequency, and replacement frequency is rather consistent throughout the entire range of simulated values (Figure 4a,d,g). For example, increasing the TMP from 2 to 9 bar results in a narrow band of consistently increasing GWP impacts (Figure 4a), suggesting that TMP dominates the other parameters throughout the range simulated. In contrast, increasing the regeneration frequency from 30 to 150 times/year results in a wide band of randomly changing GWP impacts (Figure 4b), indicating that the impact of regeneration frequency on the overall GWP impacts is overshadowed by other factors.

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Sensitivity of the eutrophication impacts to the LFM fiber lifespan is range-dependent. A narrower band in the high-impact range is seen when LFM fibers need to be replaced frequently (one replacement per 100−200 days, Figure 4f), indicating a larger impact of LFM fiber lifespan on the final eutrophication impact. In contrast, the impact of LFM fiber lifespan is dominated by the other factors when it reaches the high range (one replacement per 400−500 days), as suggested by a wider band of impact values (Figure 4f).

Different LFM treatments follow similar trends as the untreated LFMs (Figures S1, S2). The NaOH treatment reduces the sensitivity of life cycle impacts to the design and operation parameters, while the H_2O_2 treatment enhances the respective sensitivities (Figures S2, S3).

In summary, the results reveal that permeability is the most important parameter that should be targetted to further improve the life cycle environmental performance of LFMs. It can be accomplished by further improving the fouling resistance or increasing the permeate flux through chemical treatments (e.g., the H_2O_2 treatment compared in this study), as demonstrated in previous studies of LFMs and polymer membranes.[38] Between the two options, an improvement in the fouling resistance can improve all nine impact categories, whereas the chemical treatments studied so far are only effective at reducing the eutrophication impacts. In addition, with chemical treatments, the trade-off between environmental and technical performance needs to be balanced carefully. While chemical treatments can improve the environmental performance of LFMs, they also change the technical performance (i.e., size cutoff).[4,39]

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.0c01306.

Overview of LFM preparation and use; life cycle inventory data; simulation results for NaOH- and H_2O_2-treated LFMs (PDF)

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Notes

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