Electronic Supplementary Material (ESI) for Environmental Science: Water Research & Technology.

Supplementary Appendix

Enhancing Nitrification Fluxes and Nitritation Efficiencies in MABRs: A Modeling Study

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The following is included as a supplementary appendix for this paper:

Past Research

Past experimental studies were reviewed to seek trends in MABR behavior. Results are summarized in Table S1 and Figure S1. As shown in Figure S1a, fluxes generally increase with increasing bulk NH_4^+. Greater air supply pressures also result in higher fluxes, especially at higher bulk NH_4^+ concentrations (Figure S1b). An increasing concentration of COD in the bulk liquid decreases the nitrification flux, as seen in Figure S1c. Note that each curve in Figure S1 is from a different study, where the air supply pressures, membrane materials, and biofilm thicknesses may have differed. Thus, the curves are not directly comparable to each other and only can show general trends.

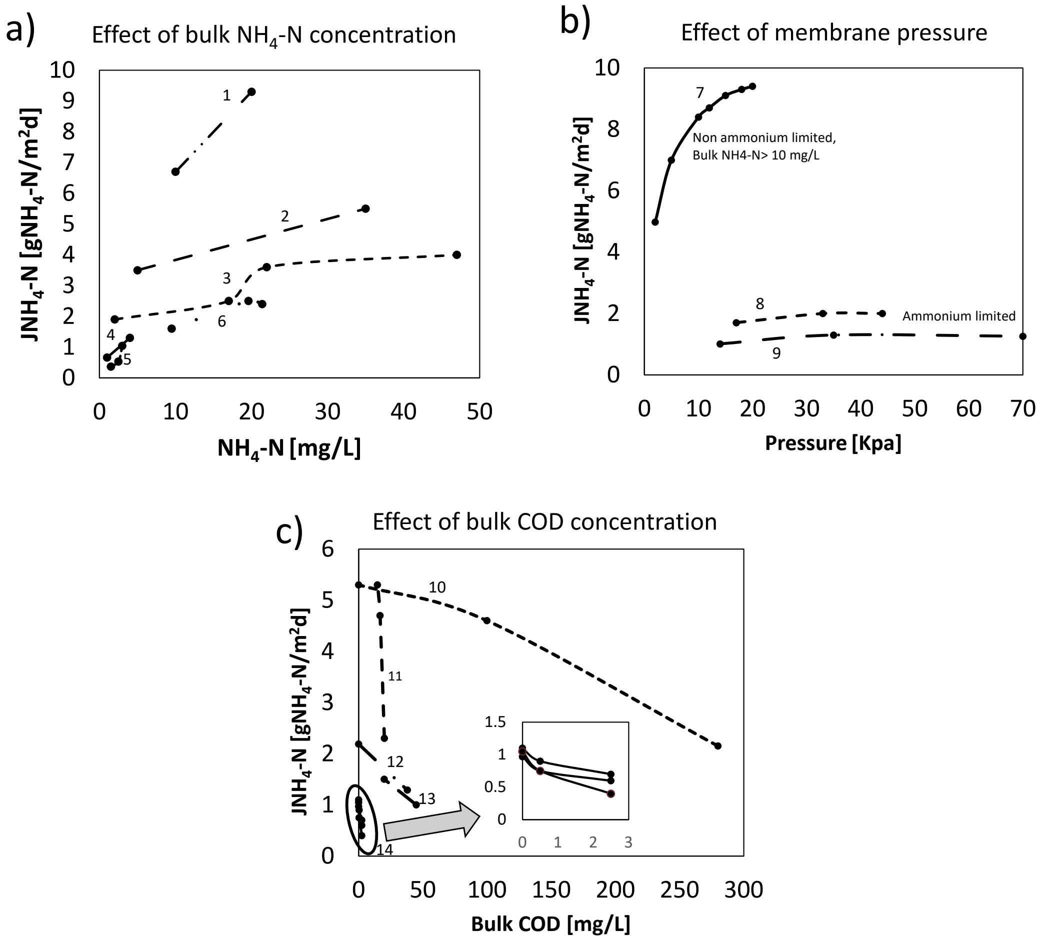


Figure S1: Selected past studies where (a) ranges of bulk NH_4^+ concentrations, (b) membrane pressures, and (c) ranges of bulk COD concentrations were studied. The number on each curve corresponds to the study listed in Table S1.

The trends observed in these experiments are discussed and explained through our simulation findings in the results and discussion section.

Table S1. Summary of relevant MABR studies

Study	Membrane type	Gas supply type and pressure (kPa)	Influent flow type	Bulk COD (mg/L)	Bulk NH_4^+ (mgN/L)	Biofilm Thickness (µm)	Bulk pH	Nitrogen loading (gN/m²-d)	Nitrificatio n flux (gN/m²-d)	Reference
1	Coal	Air, 20	CM	55 55	10 20	1000 1900	7.5-8	7.3 10.7	6.7 9.3	[1]
2	PDMS	Intermittent Air 35	Completely mixed (CM)	None added	5 35 140	540	7.2	5 13 31	3.5 5.5 9.2	[2]
3	Silicone	O_2 (Open end) Not reported	CM	None added	2 17 22 47 70 93	Not reported	5.8 6.2 6.8 7 7.3 7.5	1.97 3.6 5.7 7.7 9.5 11.8	1.9 2.5 3.6 4 4.2 4.2	[3]
4	Silicone	Air 7	CM	None added	1 4	Not reported		0.67 1.35	0.66 1.3	[4]
5	Polyvinylidene fluoride	Air 100	CM	17 13 20	1.5 2.5 3	Not reported	Not reported	0.39 0.59 1.18	0.37 0.53 1.04	[5]
6	Polypropylene flat sheet	Air 100 100 193	CM	None added	9.5 19.6 21.4	540 ± 160 560 ± 60 >1000	7.2-7.8	2.1 4.2 4.2	1.6 2.5 2.4	[6]
7	PDMS	Air 2 5 10	CM	None added	49 33 14	600 ± 12	7.5-8.3	10.2 10.4 9.8	4.98 7 8.4	[7]
8	Nonporous silicone	O_2 17 33 45 62 54 49 47	Upflow MABR	None added	14 3 4.8 11 11 10	Not reported	7-7.5	2.13 2.13 2.13 4.2 3.4 2.8 2.4	1.7 2 1.9 3.5 2.8 2.4 2.2	[8]

Study	Membrane type	Gas supply type and pressure (kPa)	Influent flow type	Bulk COD (mg/L)	Bulk NH_4^+ (mgN/L)	Biofilm Thickness (µm)	Bulk pH	Nitrogen loading (gN/m^2-d)	Nitrificatio n flux (gN/m^2-d)	Reference
9	Mitsubishi, composite	Air, closed 14 35 70	CM	None added	2.3 2.05 1.78 1.82	80 100 100 120	Not reported	33	0.74 1.01 1.3 1.26	[9]
10	PDMS	O_2 20	CM	280 100 0	720 440 500	Not reported	7.5-7.8	12.85	2.15 4.62 5.31	[10]
11	ZeeLung™	Air, (Throttled) 35	CM	20.1 16.7 14.7	22.6 14.5 14.2	Not reported	7.7 7.8 7.6	7.9 8.3 7.4	2.3 4.7 5.3	[11]
12	Coal	Air 25	CM	0 38	0.5 25	Not Reported 800-1800	7.5-8	2.21 2.33	2.19 1.29	[12]
13	PVDF	Air 100	CM	45 20	37 20	Not reported	6.7 7.2	2.6 2.6	1 1.5	[13]
14	Mitsubishi, composite	Air, closed Pressure not reported	CM	0 0-0.5 0.5-2.5 0 0-0.5 0.5-2.5 0 0-0.5 0.5-2.5	6.7 9.1 11.5 11.4 13.3 14.7 3.7 6.4 9.48	120 ± 25	Not reported	1.65 1.65 1.65 2.24 2.24 2.24 1.46 1.46 1.46	1.1 0.9 0.7 0.97 0.75 0.6 1.05 0.75 0.4	[14]

Effect of intra-membrane O_2 pressure

Increasing the intramembrane air pressure for a MAB increases the J_{NH4} but the magnitude depends on if the biofilm is ammonium limited, DO limited, or both (Figure S2).

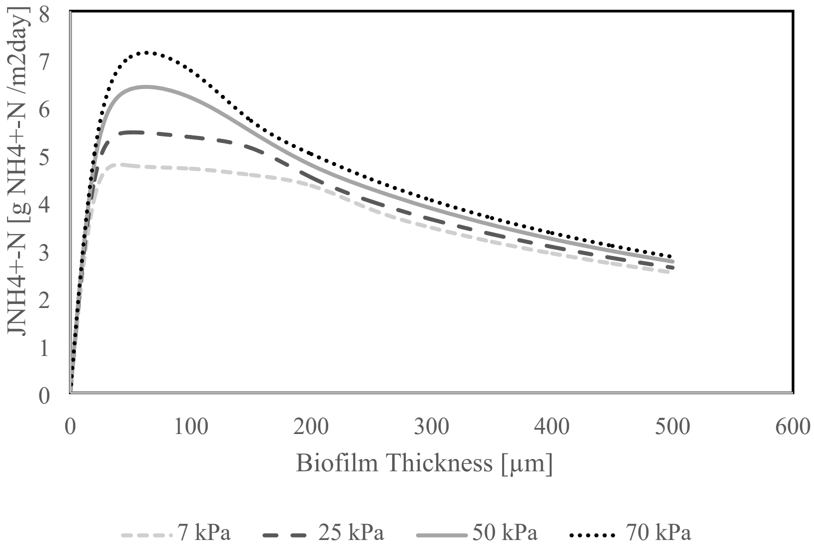


Figure S2. Effect of intramembrane air pressures of 7, 25, 50 and 70 kPa pressure on ammonium oxidation fluxes when the NH_4^+ concentration is 10 mgN/L.

Effect of Bulk BOD

Nitrifiers can grow in the inner biofilm, near the membrane surface, while BOD is consumed in the external biofilm with the nitrate and/or nitrite produced by the nitrifiers (Figure S3c and d).

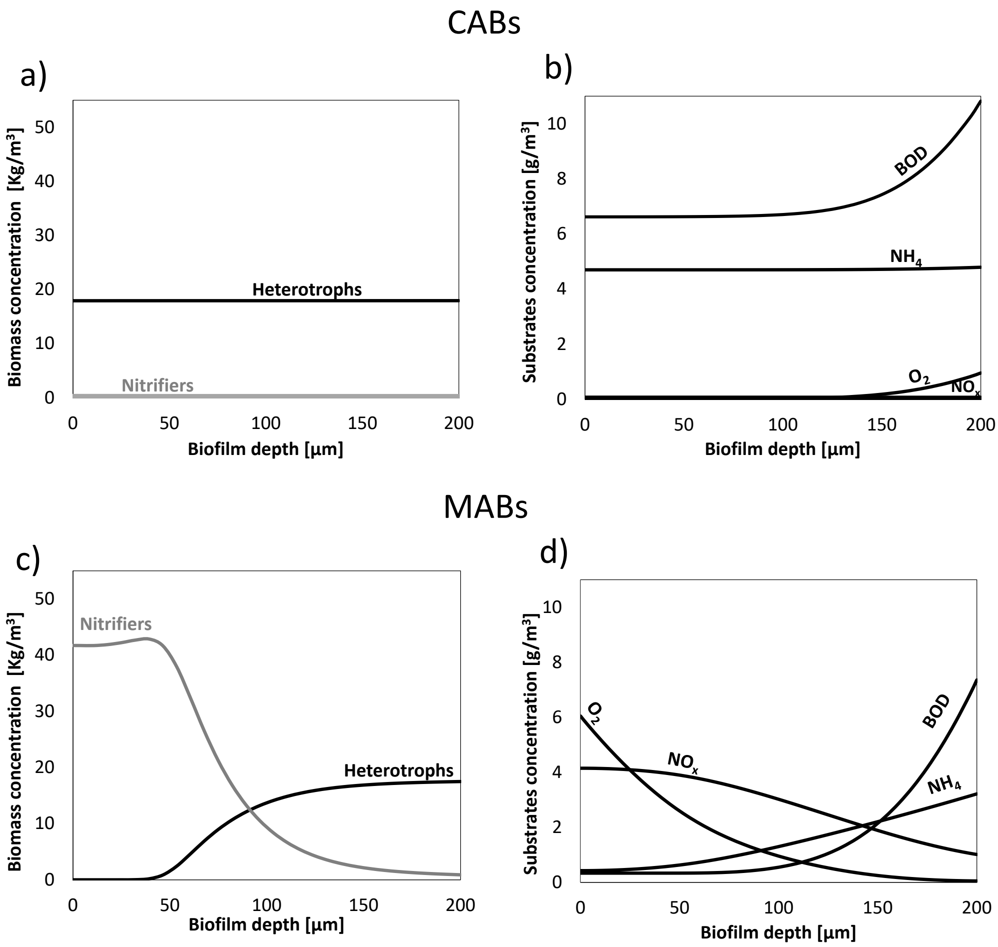


Figure S3: Model predicted biomass and substrate concentration profiles for CABs and MABs. Both systems are simulated with bulk NH_4^+ and BOD concentrations of 5 mgN/L and 20 mg/L, respectively. For the MAB, the intra-membrane relative air pressure is 40 kPa, for the CAB the bulk DO concentration is 3 mg/L. An LDL of 100 µm is considered in all the simulations (LDL profile not shown in the graphs). "Biofilm depth" on the x axis refers to the distance from the attachment surface.

Sensitivity Analyses

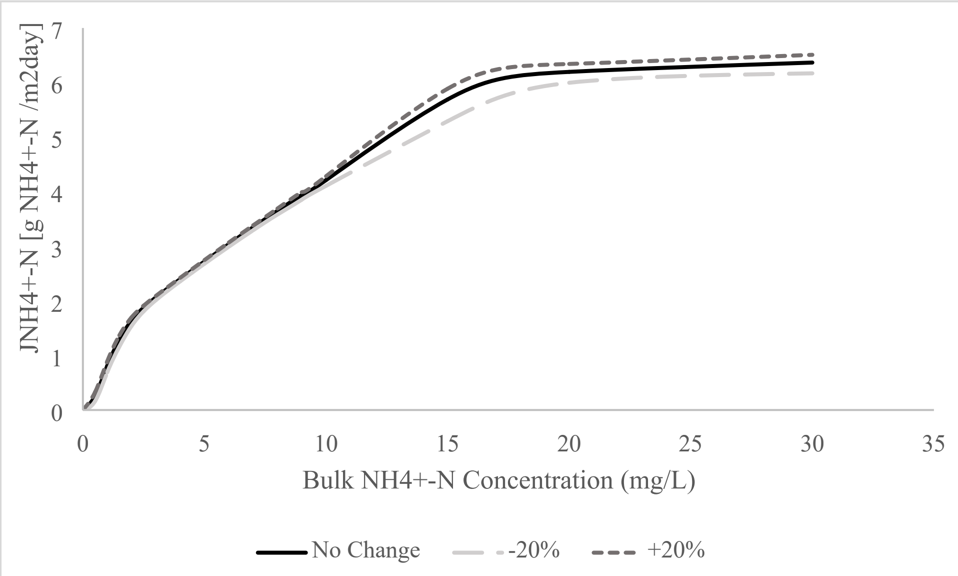
We performed a sensitivity analysis on the how different biokinetic parameters would impact the nitrification flux and the nitritation percentage by changing each parameter by positive and negative 20%. The effects on the ammonium removal fluxes are seen in Table S2 below.

Table S2. The effect of changes in different biokinetic parameters on the ammonium removal fluxes.

Parameter	**% change**	**% change in ammonium removal flux at 5 mg/L bulk NH_4^+**	**% change in ammonium removal flux at 15 mg/L bulk NH_4^+**
μ_{max} (AOB)	-20%	-1.51%	-7.01%
	20%	0.96%	3.45%
μ_{max} (NOB)	-20%	0.42%	1.57%
	20%	-0.32%	-3.73%
b (AOB)	-20%	0.99%	1.15%
	20%	-1.01%	-1.26%
b (NOB)	-20%	-0.24%	-0.43%
	20%	0.24%	0.39%
K_{O2} (AOB)	-20%	0.15%	1.07%
	20%	-0.15%	-1.10%
K_{O2} (NOB)	-20%	-0.24%	-2.11%
	20%	0.20%	1.24%
K_{NH4} (AOB)	-20%	0.98%	2.47%
	20%	-0.15%	-1.10%
K_{NH4} (NOB)	-20%	0.00%	-0.02%
	20%	0.01%	0.02%
K_{NO2} (AOB)	-20%	0.00%	0.00%
	20%	0.00%	0.00%
K_{NO2} (NOB)	-20%	-0.33%	-1.02%
	20%	0.29%	0.76%
K_{NO3} (AOB)	-20%	-0.01%	-0.01%
	20%	0.01%	0.01%
K_{NO3} (NOB)	-20%	0.01%	0.00%
	20%	-0.01%	-0.01%

Since the μ_{max} for AOB had a much larger impact on the fluxes than any of the other biokinetic parameters, we investigated how a 20% positive or negative change would change the trends that the model found. We compared the removal fluxes across different bulk ammonium concentrations (Figure S4a) and biofilm thicknesses (Figure S4b).

(a)



(b)

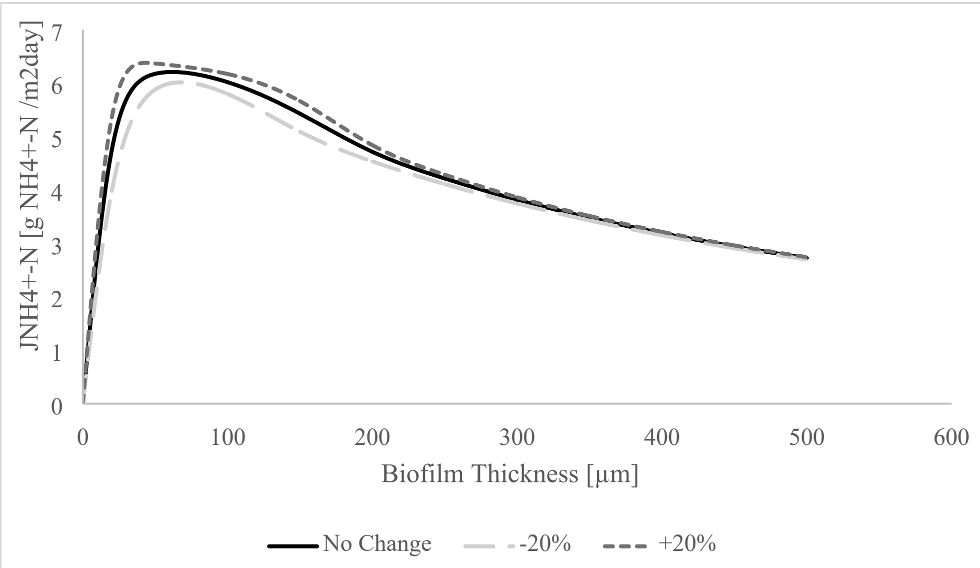


Figure S4. Effect of a positive or negative 20% change in μ_{max} for AOB on ammonium removal fluxes across different (a) bulk ammonium concentrations and (b) biofilm thicknesses.

As can be seen in Figure S4, the removal fluxes were very similar for a higher or lower μ_{max} for AOB and the trends were the same. Therefore, even if the values used in this model were not exact, the trends and findings would still be valid.

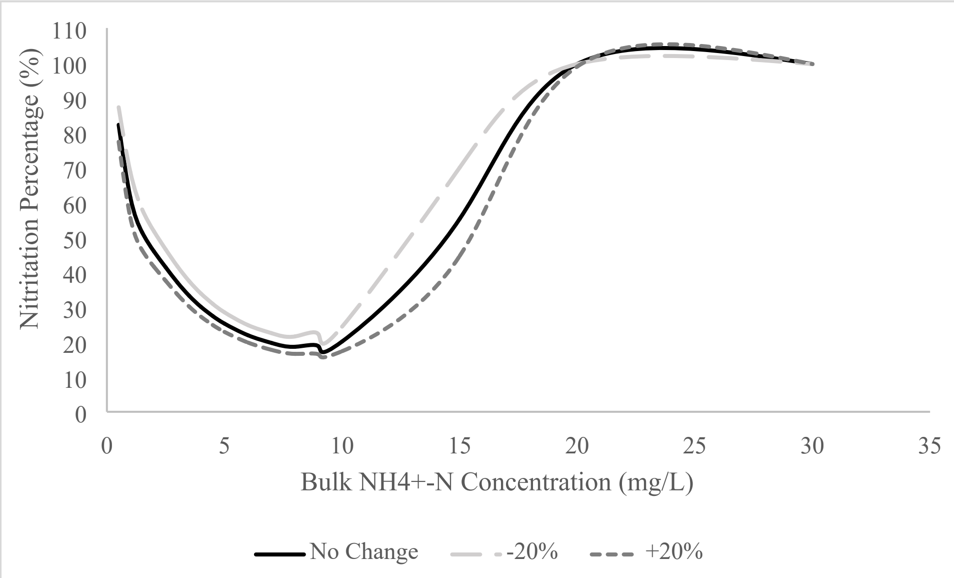
A similar analysis was done with nitritation percentages, as shown in Table S3.

Table S3. The effect of changes in different biokinetic parameters on the nitritation percentages.

Parameter	% change	% change in nitritation percentage at 5 mg/L	% change in nitritation percentage at 15 mg/L
μ_{max} (AOB)	-20%	-0.27%	-14.81%
	20%	0.23%	9.66%
μ_{max} (NOB)	-20%	12.81%	26.59%
	20%	-9.39%	-19.22%
b (AOB)	-20%	0.38%	3.92%
	20%	-0.27%	-4.01%
b (NOB)	-20%	-7.67%	-2.92%
	20%	7.60%	2.95%
K_{O2} (AOB)	-20%	0.54%	1.10%
	20%	-0.60%	-1.28%
K_{O2} (NOB)	-20%	-4.98%	-10.84%
	20%	4.31%	10.07%
K_{NH4} (AOB)	-20%	0.02%	7.64%
	20%	0.11%	-6.73%
K_{NH4} (NOB)	-20%	-0.23%	-0.13%
	20%	0.23%	0.13%
K_{NO2} (AOB)	-20%	0.00%	0.00%
	20%	0.00%	0.00%
K_{NO2} (NOB)	-20%	-10.04%	-9.48%
	20%	8.99%	8.17%
K_{NO3} (AOB)	-20%	-0.04%	0.00%
	20%	0.04%	0.01%
K_{NO3} (NOB)	-20%	0.23%	0.03%
	20%	-0.23%	-0.03%

The nitritation percentages were more sensitive to variations in the biokinetic parameters than the nitrification fluxes. To ensure that the trends would be maintained with different biokinetic parameters, the effect of varying the μ_{max} for NOB (Figure S5) and the K_{O2} for NOB (Figure S6) was examined.

(a)



(b)

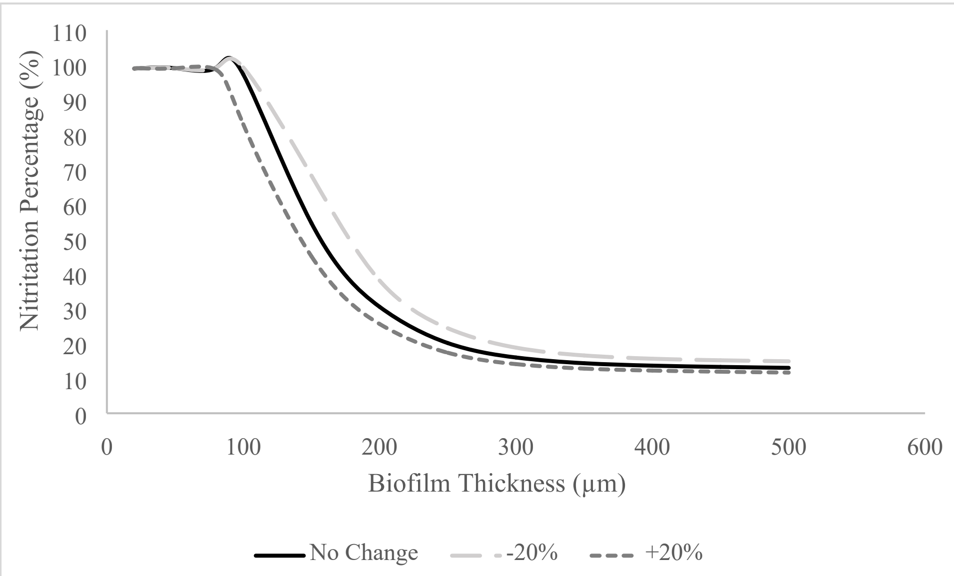
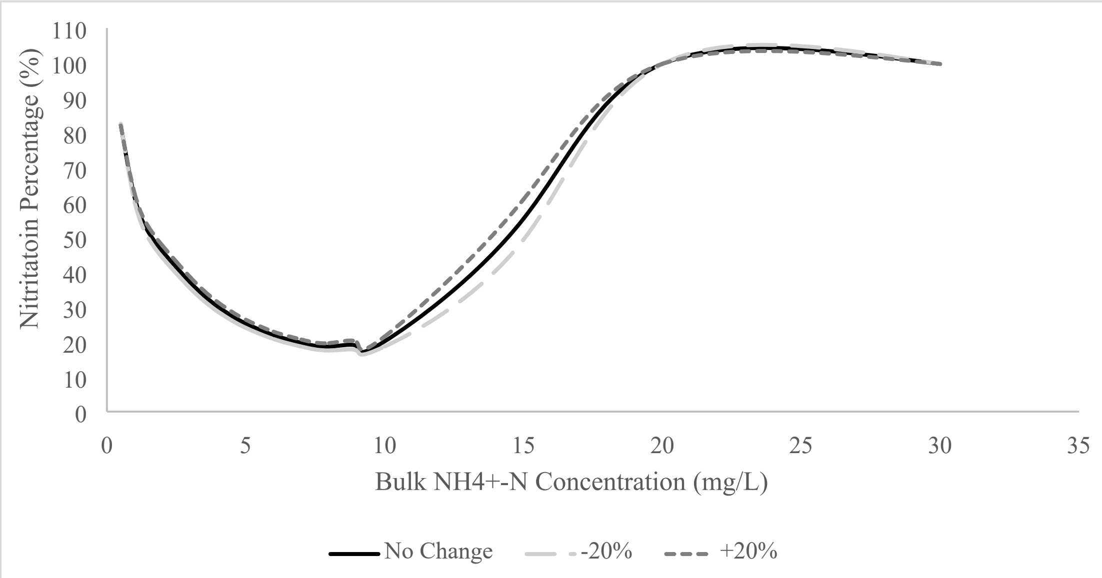


Figure S5. Effect of a positive or negative 20% change in μ_{max} for NOB on nitritation percentage across different (a) bulk ammonium concentrations and (b) biofilm thicknesses.

Though the μ_{max} for NOB resulted in the largest differences in nitritation percentages, a 20% positive or negative change did not change any of the trends. A similar procedure was done for the K_{O2} for NOB.

(a)



(b)

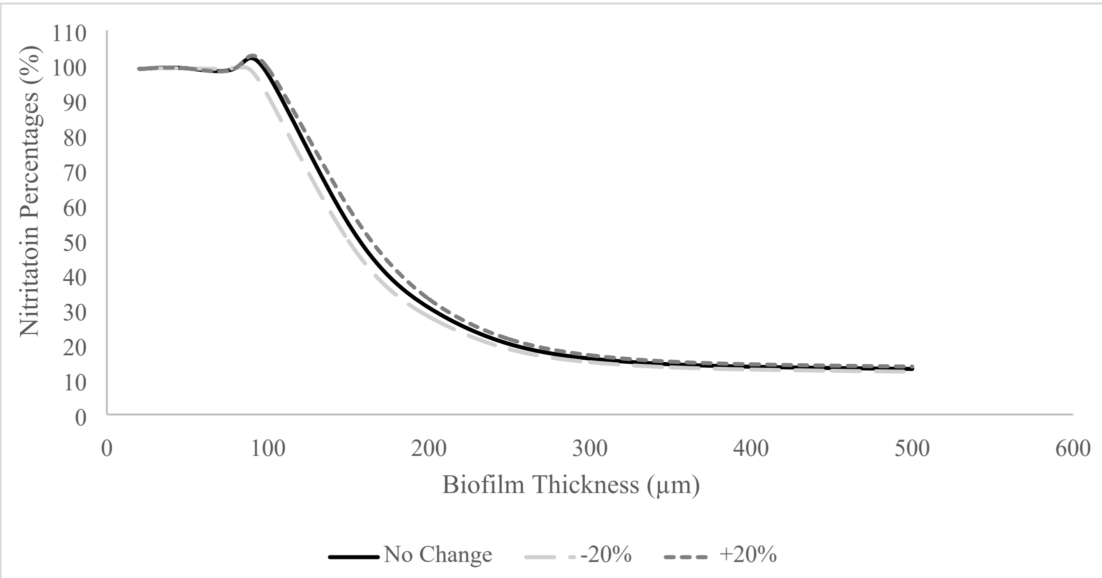


Figure S6. Effect of a positive or negative 20% change in K_{O2} for NOB on nitritation percentage across different (a) bulk ammonium concentrations and (b) biofilm thicknesses.

As seen in Figure S6, variations in K_{O2} do not change the trends of the nitritation percentages found in these studies. Even if the parameters chosen for this study had been different, the trends and general conclusions drawn about the nitrification fluxes and nitritation percentages would remain the same.

References

1 H. Liu, F. Yang, T. Wang, Q. Liu and S. Hu, Carbon membrane-aerated biofilm reactor for synthetic wastewater treatment, *Bioprocess Biosyst Eng*, 2007, **30**, 217–224.

2 C. Pellicer-Nàcher, C. Domingo-Félez, S. Lackner and B. F. Smets, Microbial activity catalyzes oxygen transfer in membrane-aerated nitritating biofilm reactors, *Journal of Membrane Science*, 2013, **446**, 465–471.

3 Y.-L. Hsieh, S.-K. Tseng and Y.-J. Chang, Nitrification using polyvinyl alcohol-immobilized nitrifying biofilm on an O2-enriching membrane, *Biotechnology Letters*, 2002, **24**, 315–319.

4 M. R. Augusto, P. R. Camiloti and T. S. O. de Souza, Fast start-up of the single-stage nitrogen removal using anammox and partial nitritation (SNAP) from conventional activated sludge in a membrane-aerated biofilm reactor, *Bioresource Technology*, 2018, **266**, 151–157.

5 Y. Li and K. Zhang, Pilot scale treatment of polluted surface waters using membrane-aerated biofilm reactor (MABR), *Biotechnology & Biotechnological Equipment*, 2018, **32**, 376–386.

6 J. W. Shanahan and M. J. Semmens, Alkalinity and pH effects on nitrification in a membrane aerated bioreactor: An experimental and model analysis, *Water Research*, 2015, **74**, 10–22.

7 R. Wang, F. Xiao, Y. Wang and Z. Lewandowski, Determining the optimal transmembrane gas pressure for nitrification in membrane-aerated biofilm reactors based on oxygen profile analysis, *Appl Microbiol Biotechnol*, 2016, **100**, 7699–7711.

8 X. Li, S. Sun, B. D. Badgley, S. Sung, H. Zhang and Z. He, Nitrogen removal by granular nitritation–anammox in an upflow membrane-aerated biofilm reactor, *Water Research*, 2016, **94**, 23–31.

9 L. S. Downing and R. Nerenberg, Effect of oxygen gradients on the activity and microbial community structure of a nitrifying, membrane-aerated biofilm, *Biotechnology and Bioengineering*, 2008, **101**, 1193–1204.

10 R. Wang, X. Zeng, Y. Wang, T. Yu and Z. Lewandowski, Two-step startup improves pollutant removal in membrane-aerated biofilm reactors treating high-strength nitrogenous wastewater, *Environmental Science: Water Research & Technology*, 2019, **5**, 39–50.

11 C. Duvall, The University of Guelph, 2017.

12 H. Liu, F. Yang, S. Shi and X. Liu, Effect of substrate COD/N ratio on performance and microbial community structure of a membrane aerated biofilm reactor, *Journal of Environmental Sciences*, 2010, **22**, 540–546.

13 H.-L. Tian, J.-Y. Zhao, H.-Y. Zhang, C.-Q. Chi, B.-A. Li and X.-L. Wu, Bacterial community shift along with the changes in operational conditions in a membrane-aerated biofilm reactor, *Appl Microbiol Biotechnol*, 2015, **99**, 3279–3290.

14 L. S. Downing and R. Nerenberg, Total nitrogen removal in a hybrid, membrane-aerated activated sludge process, *Water Research*, 2008, **42**, 3697–3708.