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# Autotrophic nitrogen removal in membrane-aerated biofilms: Archaeal ammonia oxidation versus bacterial ammonia oxidation

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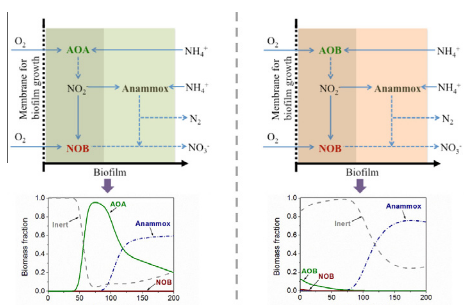
## HIGHLIGHTS

- System performance of AOA–Anammox MABR was assessed using mathematical modeling.
- AOA–Anammox MABR shows higher TN removal/lower oxygen supply than AOB–Anammox MABR.
- AOA–Anammox MABR shows wider operating window for high-level TN removal.
- This study provides first insight on design and operation of novel AOA–Anammox MABR.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Recent discovery of ammonia-oxidizing archaea (AOA) not only substantially improved our understanding of the global nitrogen cycle, but also provided new possibilities for nitrogen removal from wastewater. In particular, compared to ammonia-oxidizing bacteria (AOB), the high ammonia oxidation under oxygen-limited conditions driven by AOA is potentially more suitable for autotrophic nitrogen removal in a single-stage membrane aerated biofilm reactor (MABR) through coupling with anaerobic ammonia oxidation (Anammox). In this work, mathematical modeling is applied to assess the system performance and associated microbial community structure of an AOA–Anammox MABR under low- (30 mg N $L^{-1}$) and high-strength (500 mg N $L^{-1}$) ammonium conditions, with a side-by-side comparison to an AOB–Anammox MABR system under the same conditions. Results demonstrate that both ammonium surface loading (or hydraulic retention time) and oxygen surface loading significantly affect the system performance. In contrast to AOB–Anammox system, the AOA–Anammox MABR shows higher total nitrogen (TN) removal and lower oxygen supply, with much better repression of NOB and substantially wider operating window for high-level TN removal (>80%) in terms of varied oxygen and ammonium loadings. These results provide first insights and useful information for design and operation of this novel AOA–Anammox MABR system in its potential future applications.



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## 1. Introduction

Nitrogen removal is one of the crucial processes in wastewater treatment. Conventional biological nitrogen removal (BNR) from wastewater consists of nitrification by ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB) followed by denitrification, which cannot satisfy the development requirements for the next generation wastewater treatment processes, such as to enhance energy recovery from organics in wastewater and to reduce the energy consumption/carbon footprint for nitrogen removal [1]. Autotrophic nitrogen removal system has been developed in which, under oxygen-limited conditions, AOB convert ammonia to nitrite, which provides electron acceptor to anaerobic ammonia oxidation (Anammox) bacteria to oxidize the remaining ammonia forming nitrogen gas [2–5].

Compared to the conventional nitrification and denitrification process, this autotrophic process has a lower oxygen demand (hence less aeration and energy consumption) and does not require external carbon source [6]. Such autotrophic system has successfully been implemented in full-scale application [7,8]. Approximately 100 full-scale autotrophic partial nitritation/anammox systems have been installed worldwide for side-stream (75%) and main-stream (25%) ammonium treatment [9], with 88% of these plants being operated as single-stage systems which would reduce the space as well as decrease both the capital and operational costs compared to two-stage reactor configurations. However, it is typically difficult to achieve high levels of nitrogen removal in one-stage autotrophic systems, because a high dissolved oxygen (DO) concentration inhibits the anammox bacteria and induces the proliferation of NOB while a low DO inhibits the activity of AOB [10,11]. Further, compared to more widely applied autotrophic treatment for high-strength ammonium, the low-strength ammonium treatment (e.g., main-stream condition) is being explored at an infancy stage and one of the main challenges is the repression of NOB [11,12].

It has been demonstrated that autotrophic ammonia oxidation is not conducted exclusively by bacteria (i.e., AOB), and ammonia-oxidizing archaea (AOA) are ubiquitous in various natural environments [13–15] and wastewater treatment systems [16–18]. Recent discovery of AOA not only substantially improved our understanding of the global nitrogen cycle, but also provided new possibilities for BNR from wastewater [19,20]. In contrast to AOB, AOA often thrive at DO levels of ca. 0.1 mg/L and can achieve higher ammonia oxidation under oxygen-limited conditions [21]. AOA are therefore likely a better partner with anammox than AOB in autotrophic nitrogen removal process. In natural systems such as marine or soil, substantial experiments have proven AOA could provide nitrite and create anoxic microenvironments for anammox bacteria with oxygen consumption [22]. Thus, an autotrophic nitrogen removal process driven by AOA and Anammox could potentially be an attractive alternative to AOB–Anammox system for achieving complete autotrophic nitrogen removal.

Due to the slow growth rates of AOA and Anammox, biomass retention is crucial for high-rate performance. Biofilms can retain microorganisms with very slow growth kinetics, and biomass can be naturally accumulated in the biofilm at different depths. Particularly, the membrane aerated biofilm reactor (MABR) has evolved in recent years [23–25] and been proved to be suitable for one-stage autotrophic nitrogen removal process [26–29]. In such system, oxygen is supplied through a gas-permeable membrane that also serves as biofilm support. The merits of such a system lie in the high and efficient oxygen transfer through the membrane and also in the potential for a more amenable control strategy due to separation of oxygen and ammonium fluxes. For example, partial nitrification and completely nitrogen removal were successfully achieved in a single MABR, with a max removal rate of 0.77 kg-N/m$^3$/d and 88.5% total nitrogen (TN) removal efficiency [27].

Mathematical modeling of wastewater treatment processes is of great importance toward a full understanding of the complex system and the optimization of its practical application [30–34]. Although AOA have several characteristics potentially rendering AOA–Anammox more suitable for AOB–Anammox, such new coupling systems have not been explored yet. In this study, we carry out a model-based assessment of the performance of a novel autotrophic nitrogen removal technology based on AOA and Anammox in one-stage MABR, for both low-strength and high-strength ammonium treatment, with a side-by-side comparison to an AOB–Anammox MABR system. The impacts of key operational parameters on removal efficiency such as oxygen surface loading ($L_{O2}$) and ammonium surface loading were investigated to provide the first insight into the role of AOA in partial nitrification MABR systems, which may help improve the design and operation of such systems for future applications.

## 2. Material and methods

### 2.1. Membrane-aerated biofilm reactors

In an MABR reactor, oxygen is supplied through a gas-permeable membrane that also serves as biofilm support (Fig. 1). Applying a counter-diffusional concept, oxygen is provided to the base of the biofilm, whereas other substrates, namely ammonium and bicarbonate, are supplied from the bulk liquid phase [35,36]. The oxygen and ammonium concentration gradients cause stratification of AOA (or AOB) and Anammox in the biofilm (Fig. 1). The simulated MABR in this work has a working volume of 1 m$^3$ with a completely mixed liquid phase. The bulk volume and biofilm surface area of the reactor are 0.96 m$^3$ and 235 m$^2$, respectively, with a surface to volume ratio of 245 m$^2$ m$^{-3}$. Gas-permeable membranes used for oxygen supply and biofilm attachment have about 0.04 m$^3$ gas volume inside the membrane lumen. Compressed air is supplied in flow-through mode to the membrane module, with the oxygen flux to the biofilm controlled through changing either the applied gas pressure or the gas flow rate into the membrane lumen. In this work, the simulated low-strength ammonium concentration is set at 30 mg N L$^{-1}$ representing main-stream condition [12] while high-strength ammonium concentration is 500 mg N L$^{-1}$ mimicking side-stream condition [9]. The influent flow rate is varied to regulate the influent ammonium surface loading, which also corresponds to hydraulic retention time (HRT) in this case.

### 2.2. Mathematical models

The kinetics and stoichiometry of the biological reaction model for the one-stage autotrophic nitrogen removal system driven by AOA and Anammox (Fig. 1A) are summarized in Tables 1 and 2. Both growth and decay processes are considered for each species, i.e., AOA, NOB, Anammox and heterotrophs. Kinetic control of all the microbial reaction rates is described by the Monod equation, with each reaction rate modeled by an explicit function considering all substrates involved. The parameters used regarding the metabolisms of AOA, NOB, Anammox and heterotrophs are listed in Table S1 in the Supporting Information (SI) with the definitions, values and units. For the one-stage autotrophic nitrogen removal system driven by AOB and Anammox (Fig. 1B), the kinetics and stoichiometry of the biological reaction model for AOB–Anammox