

Supporting information

Manuscript title: Bromate and Nitrate Bio-reduction Coupled with

Poly- β -Hydroxybutyrate Production in a Methane-based Membrane Biofilm Reactor

Authors: Chun-Yu Lai^{1, #}, Pan-Long Lv¹, Qiu-Yi Dong¹, Shi Lei Yeo¹, Bruce E.

Rittmann², He-Ping Zhao^{1, *}

1. College of Environmental and Resource Science, Zhejiang University, Hangzhou, China.

2. Biodesign Swette Center for Environmental Biotechnology, Arizona State University, P.O. Box 875701, Tempe, Arizona 85287-5701, USA.

Current address: Advanced Water Management Centre, The University of Queensland, St Lucia, Queensland 4072, Australia

* Correspondence to Dr. He-Ping Zhao. Tel (Fax): 0086-571-88982739

E-mail: zhaohp@zju.edu.cn

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Number of figures: 5

Table S1. Alpha diversity of microbial community in biofilms in Stage 1-5.

Alpha diversity parameters	Stage 1	Stage 2	Stage 3	Stage 4	Stage 5
Chao 1	264	247	233	19	196
Shannon	3.08	3.31	3.57	3.23	3.4
Simpson	0.1	0.07	0.05	0.07	0.06

Table S2. The PHB contents of the biofilms for different stages.

Stage	PHB content
Stage 1	9.6%±0.5%
Stage 2	8.4%±0.2%
Stage 4	2.3%±0.2%
Stage 6	8.9%±0.6%
Stage 7	1.9%±0.1%

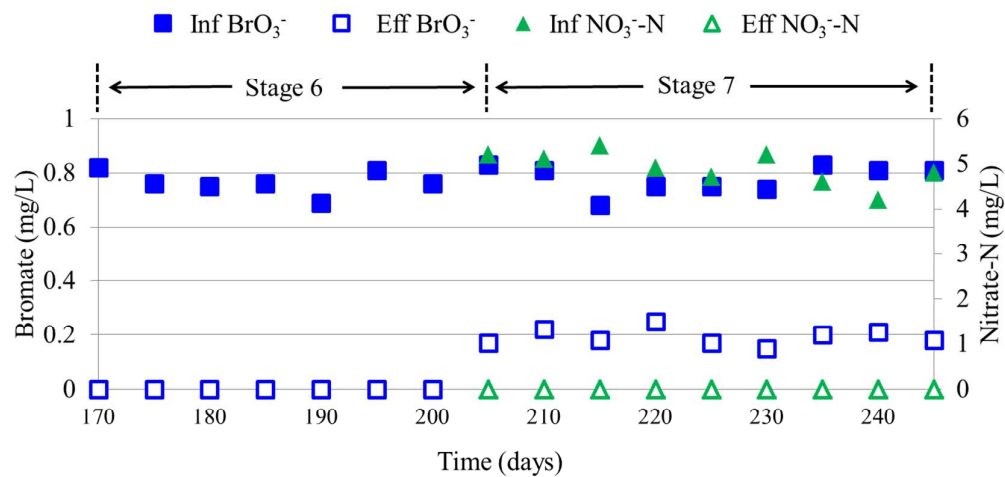


Figure S1. Performance profile for additional two stages.

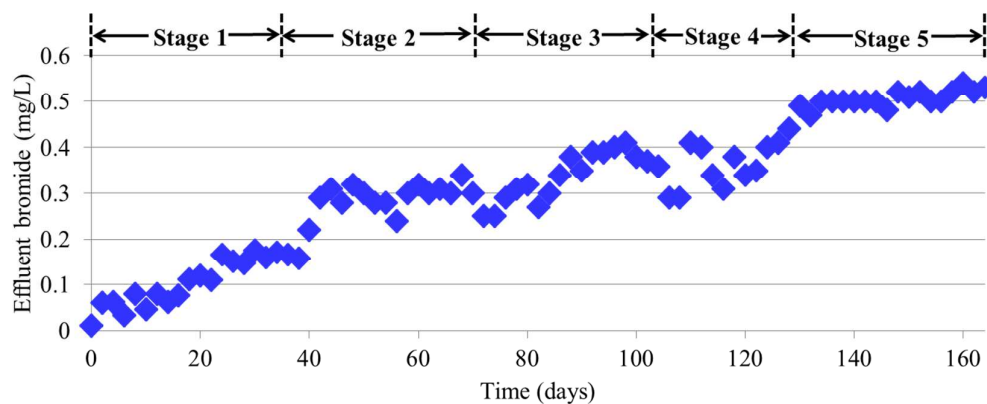


Figure S2. Bromide (Br^-) concentration in the effluent for Stage 1-5. The concentration of produced Br^- in the effluent was approximately equal to the reduced BrO_3^- , indicating that BrO_3^- was stoichiometrically reduced to Br^- . No accumulation of intermediates, such as BrO_2^- , was observed in the bulk aqueous phase.

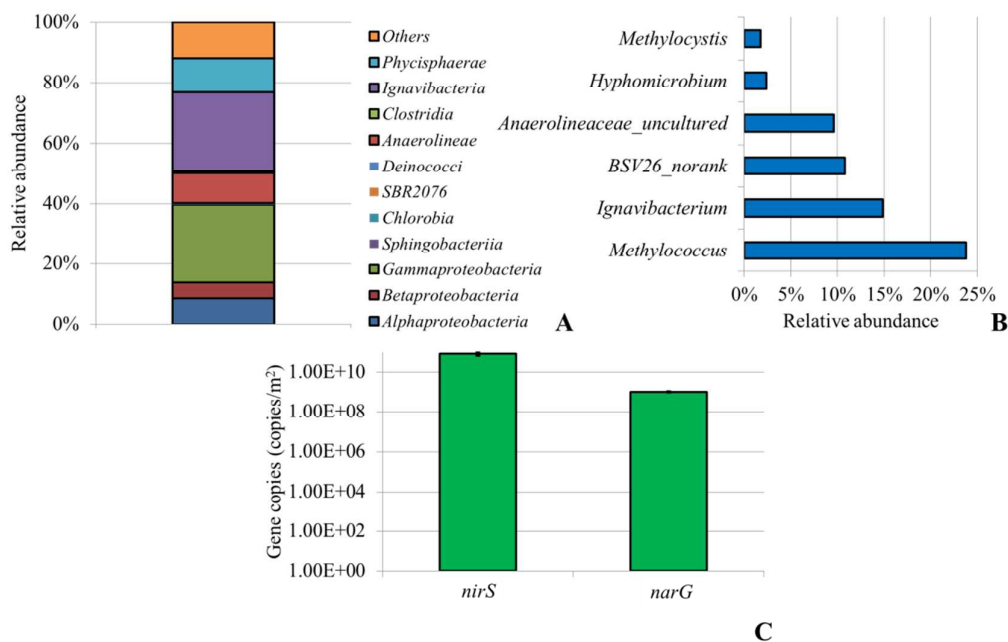


Figure S3. Dominant phylotypes at the levels of class (A) and genus (B) by Illumina sequencing, and denitrifying genes (*nirS* and *narG*) by qPCR analysis in the inoculums (C). The inoculum was taken from a mixed culture performing anaerobic oxidation of methane coupled to nitrate and perchlorate reduction for over two years. Thus, the microbial community differed from that in our previous research (Luo et al., 2015; Chen et al., 2016), as the microbial community was continually evolving. The gene copies of the inoculum were calculated as the total gene copies in the 5 mL of inoculum / the total surface area of the membrane (58 cm²)

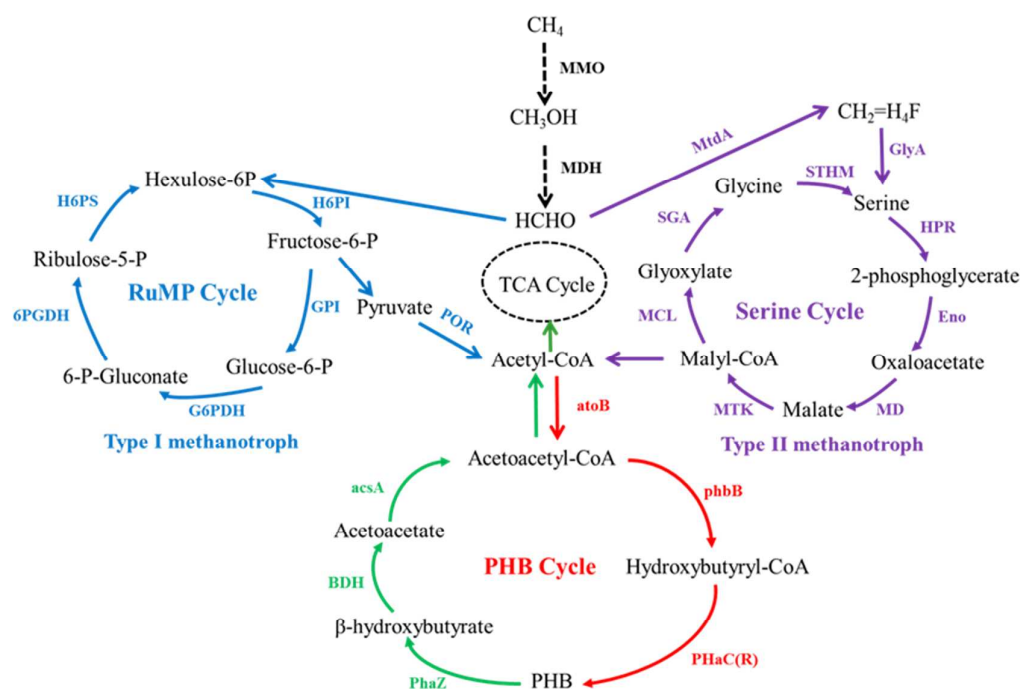


Figure S4. The methane-metabolism pathway for methanotrophs.

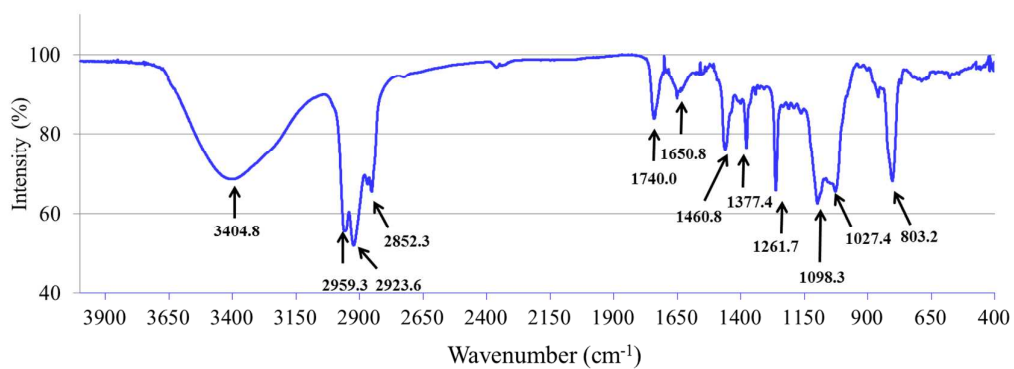


Figure S5. FTIR spectra of PHB recovered from biofilms. Key function groups of the PHB were all identified: -COOR (ester) group (carbonyl; 1650.8 cm⁻¹ and 1740.0 cm⁻¹); -OH (3404.8 cm⁻¹); C-O bonding group (1261.7 cm⁻¹ and 1098.3 cm⁻¹); asymmetric and symmetric -CH₂ stretching vibrations (1027.4 cm⁻¹, 1261.7 cm⁻¹, 1377.4 cm⁻¹, 1852.3 cm⁻¹, 2923.6 cm⁻¹ and 2959.3 cm⁻¹).

The Abbreviations for Figure S4:

Methane oxidation: MMO, methane monooxygenase; MDH, methanol dehydrogenase

Serine cycle: H4MPTP, methylene tetrahydromethanopterin pathway; MtdA, methylene tetrahydromethanopterin dehydrogenase; FDH, formate dehydrogenase; STHM, serine hydroxymethyl transferase; HPR, hydroxypyruvate reductase; MD, malate dehydrogenase; MTK, malate thiokinase; MCL, malyl coenzyme A lyase; Eno, enolase; SGA, Serine-glyoxylate aminotransferase; GlyA, Serine-hydroxymethyl transferase;

RuPM Cycle: H4MPTP, methylene tetrahydromethanopterin pathway; FDH, formate dehydrogenase; H6PI, hexulose-6-phosphate isomerase; GPI, glucose phosphate isomerase; G6PDH: glucose-6-phosphate dehydrogenase; 6PGDH: 6-phosphogluconate dehydrogenase; H6PS, hexulose-6-phosphate synthetase; POR : pyruvate oxidoreductase.

PHB cycle: atoB, Acetyl-CoA acetyltransferase; phbB, Acetoacetyl-CoA reductase

BDH: β -hydroxybutyrate dehydrogenase; PHaC(R), PHB synthetase; PhaZ, poly(3-hydroxybutyrate) depolymerase; acsA, Acetoacetyl-CoA synthetase.