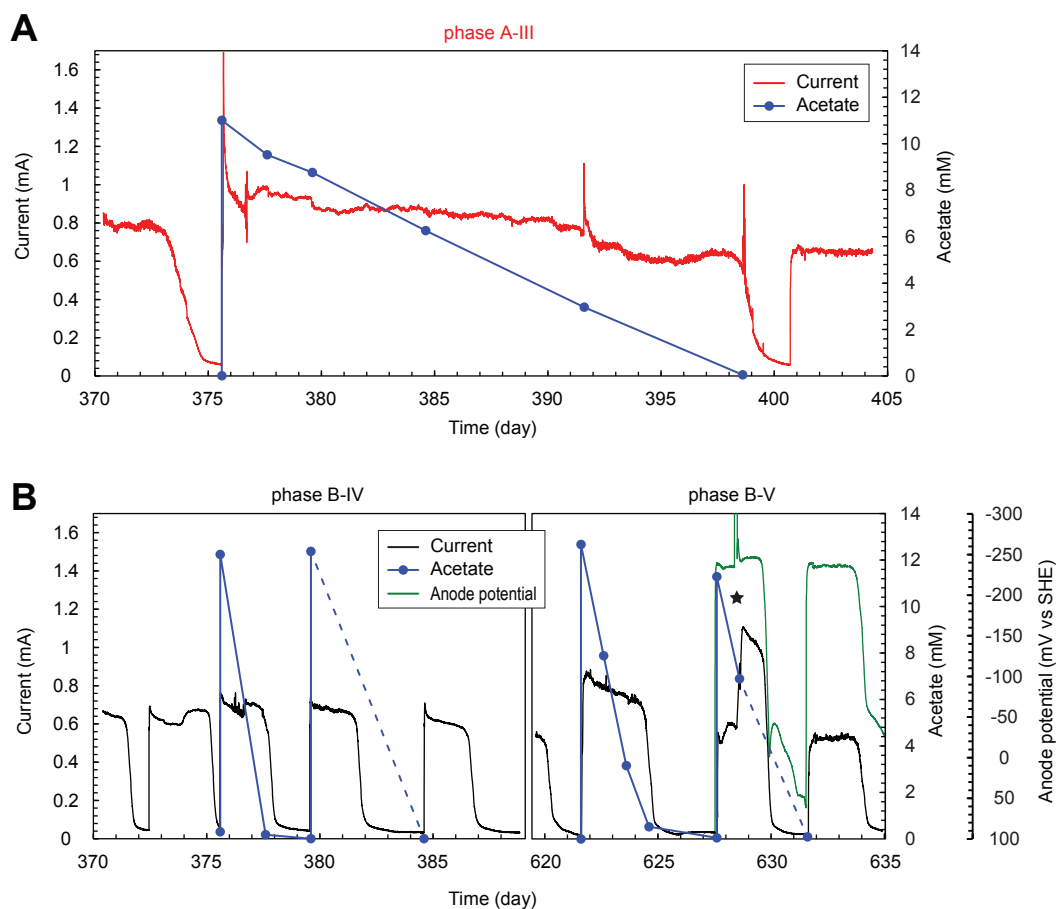
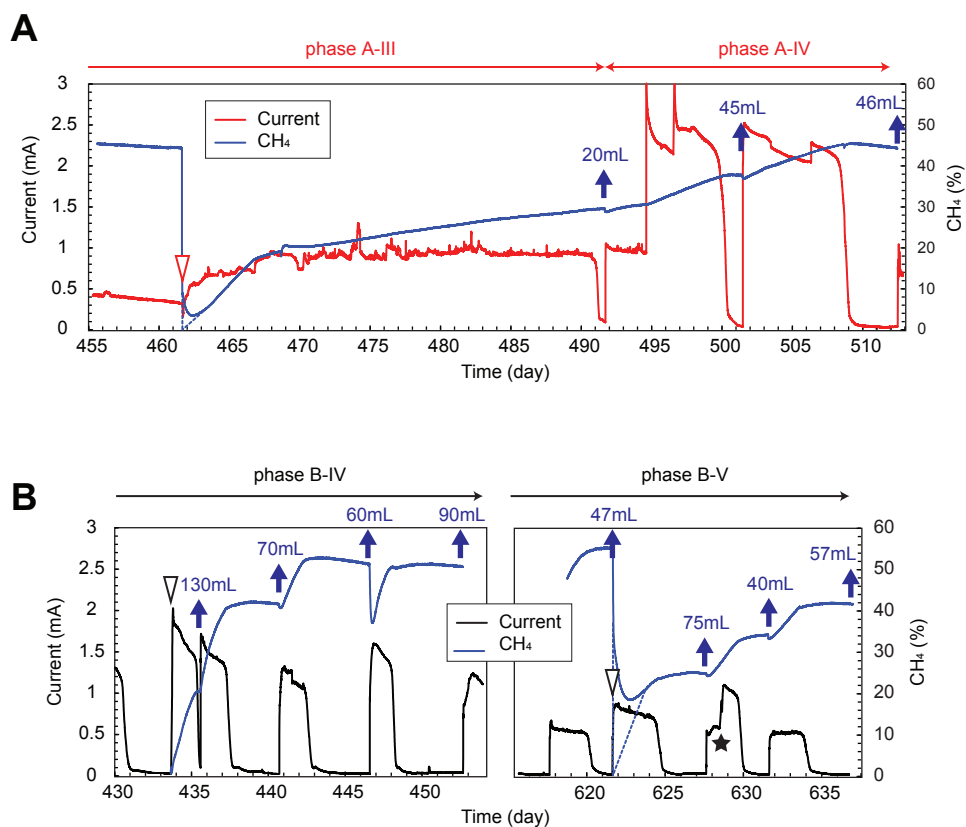




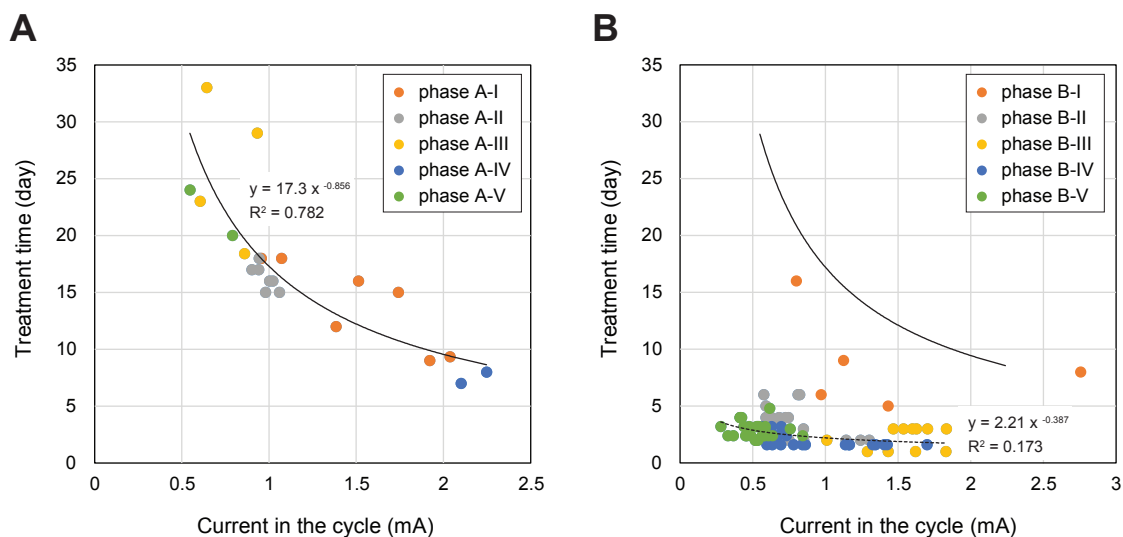
Supplementary Fig. S1 | Exemplary current and acetate consumptions in duplicated BESs.

Current generation with 600 mV of voltage application in BESs (reactor A for panel A, and reactor B for panel B) is shown as red or black lines. Acetate concentration was shown as blue filled circles. Anode potential trend (green line) is only shown for phase B-V of reactor B, and star indicates potential step voltammetry in the phase.



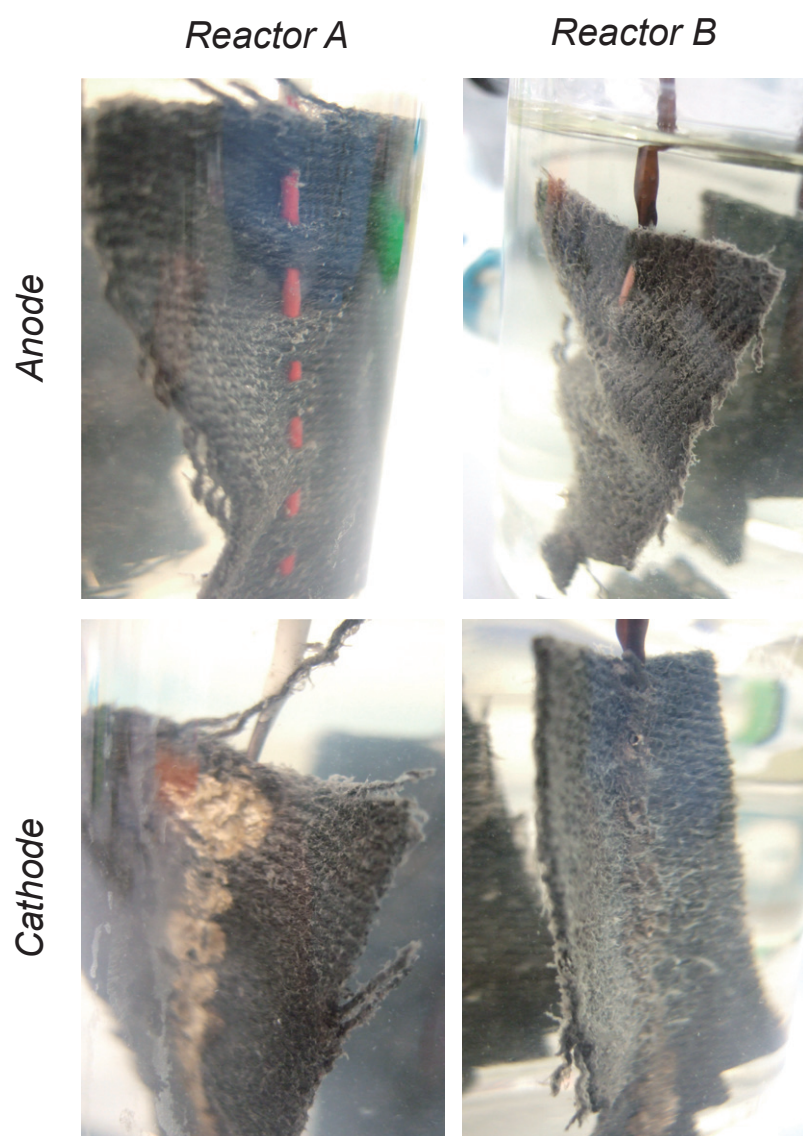
Supplementary Fig. S2 | Exemplary methane production with current consumption in duplicate BESs.

Current generation with 600 mV of voltage application in BESs (reactor A for panel A, and reactor B for panel B) is shown as red or black lines. Methane concentration (% v/v) was shown as blue lines (blue dashed line in panel B indicates estimated line, if all gas in the gas line was removed). Filled blue arrows indicate produced gas measurement from the gas pack, and the gas amounts are described above the arrows. Open arrowheads indicate exchange of formation water in the BESs. Stars indicate step voltammetry.

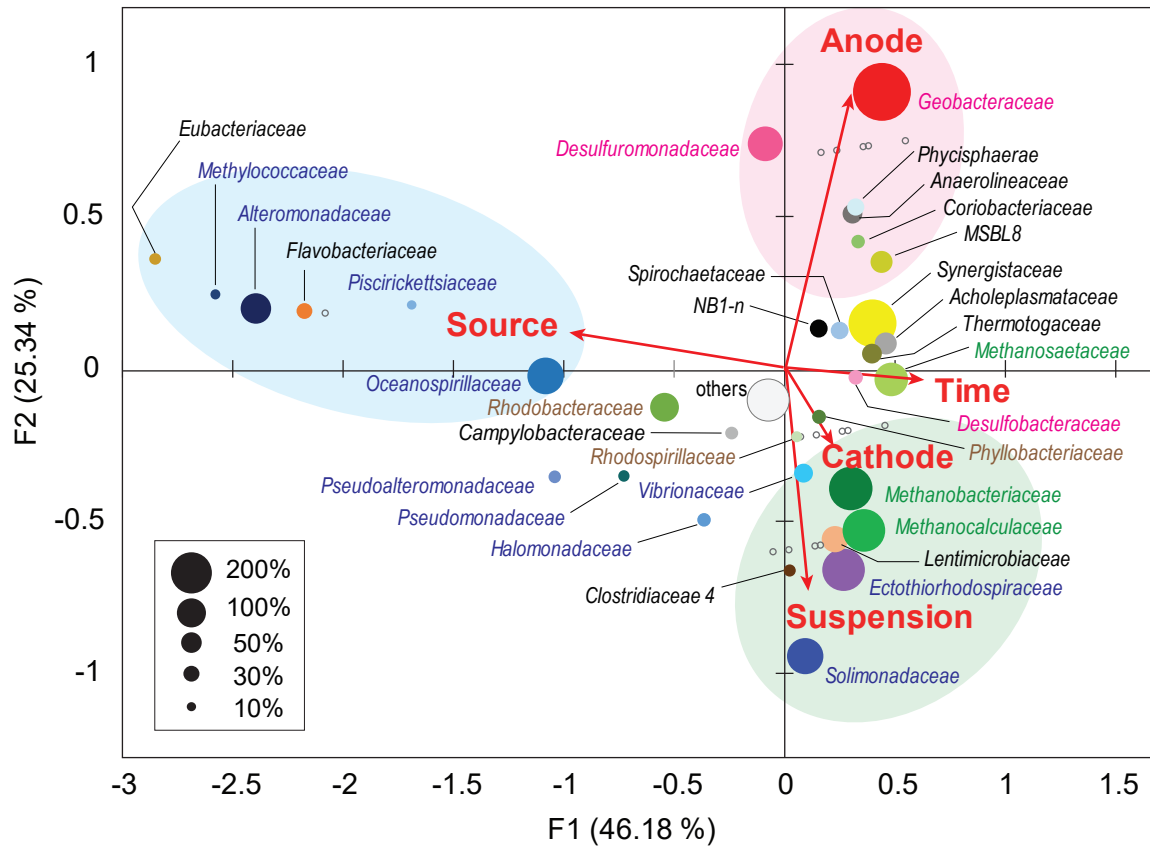


Supplementary Fig. S3 | Relationship between amounts of current and treatment time in the repeated batch cycles in the electromethanogenic BESs.

Treatment time under different current consumption is plotted in duplicate BESs (reactor A for Panel A, reactor B for Panel B). Operational phases are marked as different colors (upper right boxes). Trend line for reactor a is shown by black line, while trend line for reactor B (from phase II to phase V) is shown by dashed black line. Approximation formula and coefficient of determination are shown near lines.

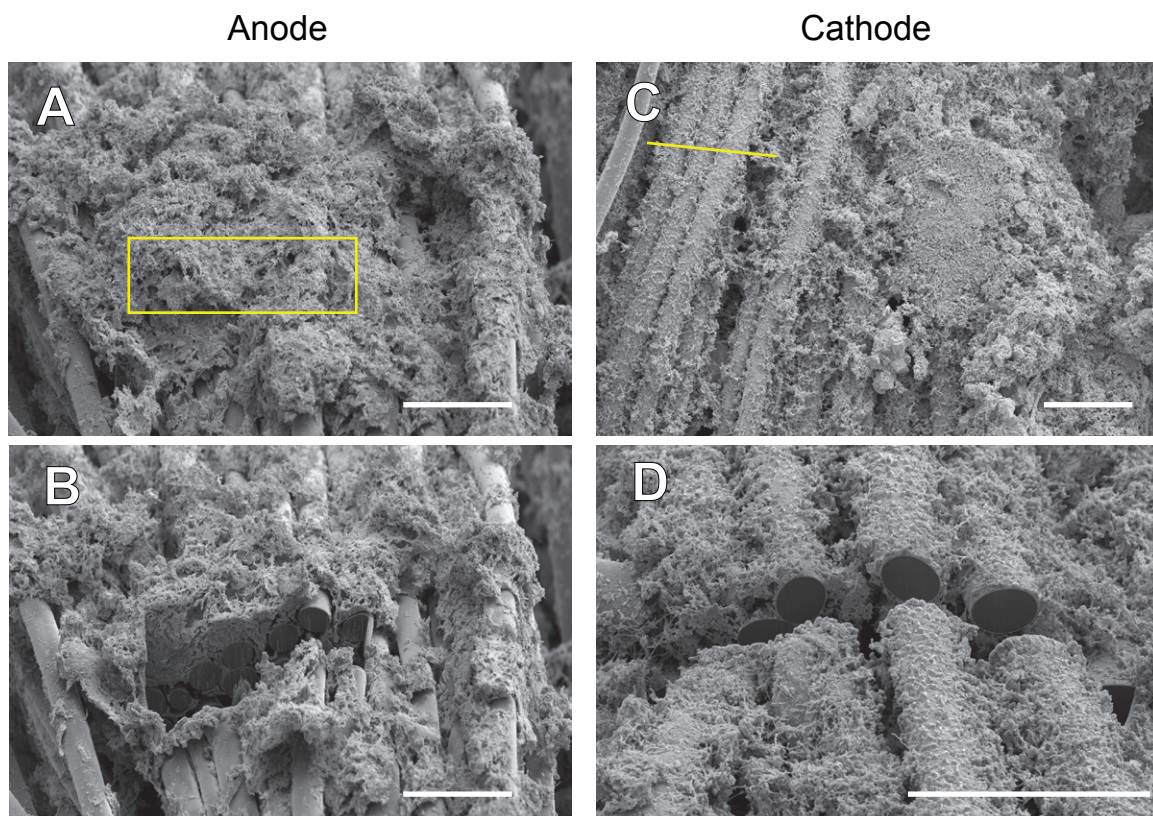


Supplementary Fig. S4 | Electrode-associated EET-active biofilms on the electrogenic anode and electromethanogenic cathode within the duplicate single-chambered BES bioreactors.



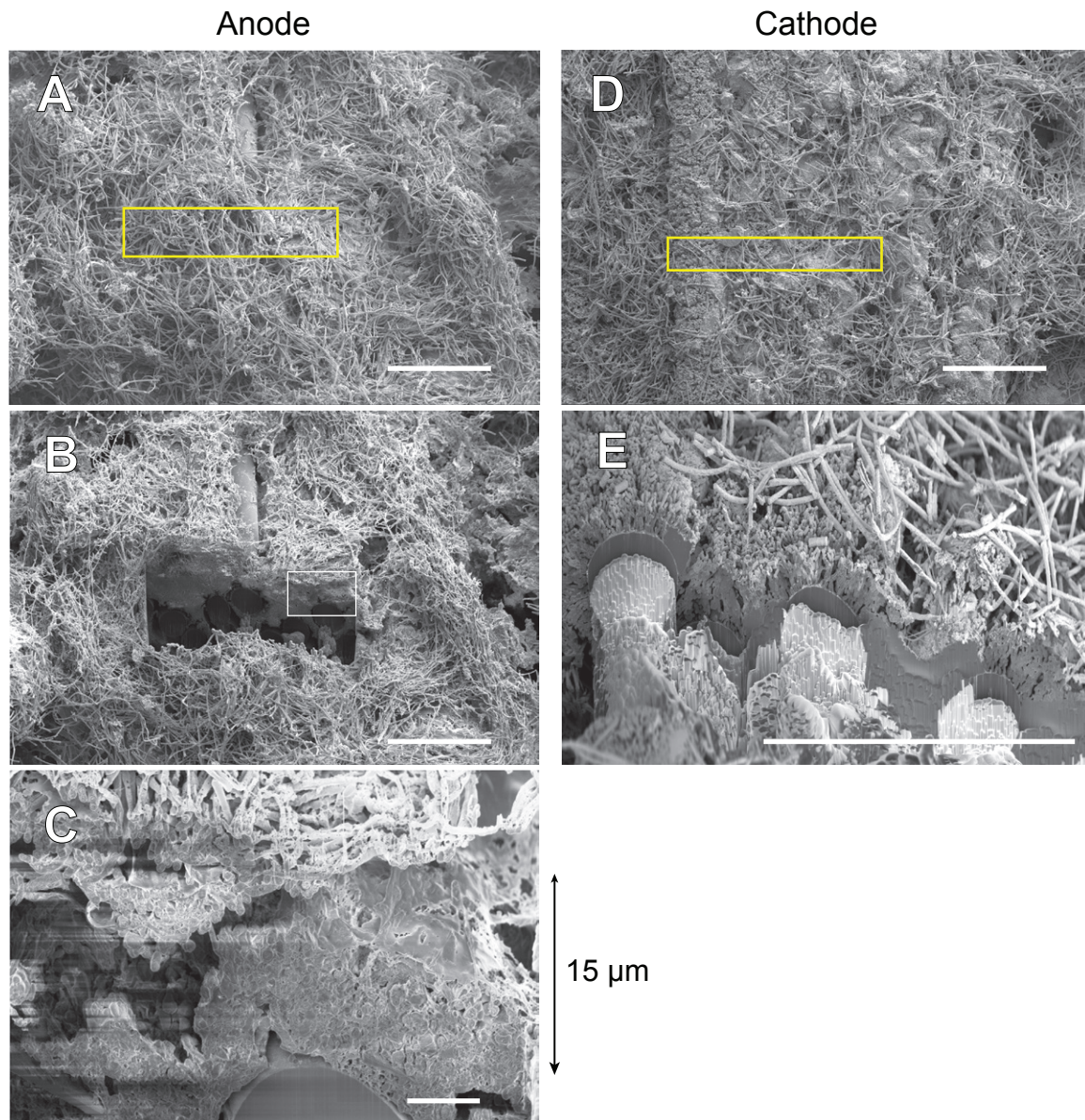
Supplementary Fig. S5 | Weighted canonical correspondence analysis (wCCA) diagram correlating microbial families and the environmental factors for all communities.

A wCCA diagram shows relationships between operational variables (red arrows) and dominant microbial families (filled bubbles) in both BES reactors. The bubble colors indicate families that described in Figure 3. Family names of the taxa are depicted near bubbles, and the names of four abundant taxa are colored as pink (*Deltaproteobacteria*), blue (*Gammaproteobacteria*), brown (*Alphaproteobacteria*) and green (*Euryarchaeota*). Bubble sizes indicate sum of relative frequencies (%) within the total 30 microbial community analyses of formation waters and both BES reactors (maximum 3,000%). Gray open circles indicate samples for microbial community analyses.



Supplementary Fig. S7 | Cross sectioning of anode and cathode biofilms in the electromethanogenic BES reactor A by using a focused ion beam (FIB).

Anode (panel AB) and cathode (panel CD) samples were collected from BES reactor A after 19-month enrichment process. Yellow rectangle in panel A shows a region digging the anode biofilm by using FIB, while yellow line in panel B shows representative location digging the cathode biofilm by using FIB. Bars = 50 μm.



Supplementary Fig. S8 | Cross sectioning of anode and cathode biofilms in the electromethanogenic BES reactor B by using a focused ion beam (FIB).

Anode (panel A-C) and cathode (panel DE) samples were collected from BES reactor B after 19-month enrichment process. Yellow rectangles in panel A and D show regions digging the biofilms by using FIB. Panel C shows thickness of the anode biofilm (white rectangle region in panel B). Bars (A, B, D, E) = 50 μm . Bars (C) = 5 μm .

Supplementary Table S1. Abundant OTUs observed in the anode biofilm, cathode biofilm, suspension or source formation waters (Top 10 OTUs for each community)

Phylum-Class		Source			A1		A2		A4		A5		B1		B2		B3		B4		B5		SUM (%)	Location	Closest relative	Accession number	
OTU ID	Family	KTG1	KTG2	KTG3	Anode	Susp.	Cathode	Anode	Susp.	Cathode	Anode	Susp.	Cathode	Anode	Susp.	Cathode	Anode	Susp.	Cathode	Anode	Susp.	Cathode					
Alphaproteobacteria																											
BRalp1	Phyllobacteriaceae	0	0	0	3	2	5	1	0	0	1	0	0	1	1	0	0	0	0	0	0	0	18	Biofilm	Cohaesibacter marisflavi DQHS21	NR_104499	
BRalp9	Phyllobacteriaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	0	3	Susp.	Mesorhizobium thiogangelicum NIOSSD020#303	KY616479	
BRalp2	Rhodospirillaceae	0	0	0	0	0	0	1	0	0	1	0	0	0	1	1	0	0	0	0	0	0	8	Biofilm	Magnetospira sp. QH-2	FO538765	
KTGalp3	Rhodobacteraceae	0	6	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	6	Source	Paracoccus fistulariae 22-5	NR_104533	
KTGalp4	Rhodobacteraceae	3	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	6	Source	Phaeobacter caeruleus DSM 24564	NR_118542	
KTGalp5	Rhodobacteraceae	0	6	0	1	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	8	Source	Iodide-oxidizing Roseovarius sp. RB-2A	AB159203	
KTGalp6	Rhodobacteraceae	2	4	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	6	Source	Sulfitobacter dubius 43-C1	LK022119	
BRalp7	Rhodobacteraceae	0	0	0	6	5	13	1	0	1	0	1	0	0	1	2	2	0	4	1	1	6	53	Cathode	Pseudoruegeria marinistellae SF-16	NR_149190	
BRalp8	Rhodobacteraceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	5	Susp.	Stappia carboxidovorans M4	NR_146031	
Deltaproteobacteria																											
BRdel1	Desulfobacteraceae	0	0	0	0	0	0	2	0	6	3	1	3	0	0	0	1	2	2	0	1	0	24	Biofilm	Desulfotignum toluenicum 4S-CH-S3-s3	MG264260	
BRdel2	Desulfuromonadaceae	0	0	0	40	9	5	3	0	0	0	0	0	1	5	1	1	0	0	1	0	0	79	Anode	Desulfuromonas sp. WB3	KM452745	
BRdel3	Desulfuromonadaceae	0	0	0	13	3	2	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	26	Anode	Desulfuromonas michiganensis BRS1	AF357914	
KTGdel5	Desulfuromonadaceae	0	0	13	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	13	Source	Desulfuromusa succinoxidans Gylac	NR_029276	
KTGdel6	Desulfuromonadaceae	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	Source	Geopsychrobacter electrodiphilus A1	NR_042768	
KTGdel7	Desulfuromonadaceae	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	Source	Pelobacter acetylenicus DSM 3246	CP015455	
BRdel8	Desulfuromonadaceae	0	0	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	0	0	0	0	15	Biofilm	Pelobacter sp. SFB93	CP015519	
BRdel9	Desulfuromonadaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	2	0	1	6	Anode	Pelobacter acetylenicus DSM 3246	CP015455	
BRdel10	Geobacteraceae	0	0	0	0	0	0	0	0	0	12	11	18	31	5	11	46	6	2	54	1	2	293	Anode	Geoalkalibacter subterraneus Red1	CP010311	
BRdel11	Geobacteraceae	0	0	0	10	7	9	30	2	4	40	0	0	0	0	0	0	0	0	0	0	0	102	Anode	Geoalkalibacter subterraneus Red1	CP010311	
BRdel12	Desulfovibrionaceae	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	2	Cathode	Pseudodesulfobacterium aespoensis Aspo-2	NR_074871	
Gammaproteobacteria																											
BRgam1	Solimonadaceae	0	0	0	3	30	3	0	27	1	1	37	0	0	0	0	4	34	3	0	5	0	154	Susp.	Solimonas terrae KIS83-12	NR_125692	
BRgam2	Halomonadaceae	0	0	0	2	11	3	0	0	0	0	0	0	1	2	1	0	0	0	0	0	0	19	Susp.	Halomonas alkaliphila M11	MF619984	
BRgam3	Vibrionaceae	0	0	0	5	8	28	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	42	Cathode	Vibrio alginolyticus SNSAB08	MH213144	
BRgam4	Ecotiorhodospiraceae	0	0	0	2	4	4	1	19	3	4	11	2	0	1	3	6	36	15	20	1	8	212	Susp.	Thioalbus denitrificans Su4	NR_122087	
BRgam5	Ecotiorhodospiraceae	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0	0	0	0	0	0	0	3	Susp.	Sulfurivermis fontis	LC225746	
KTGgam6	Methylococcaceae	2	8	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	12	Source	Methylochromium japonense NI	NR_043450	
BRgam7	Alteromonadaceae	0	0	0	1	5	3	0	0	0	0	0	0	1	1	1	0	0	1	0	0	0	18	Susp.	Marinobacter hydrocarbonoclasticus KS-An-U5	LT222322	
KTGgam8	Alteromonadaceae	13	20	55	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	89	Source	Marinobacter antarcticus HEX61	KY508320	
KTGgam9	Piscirickettsiaceae	2	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	5	Source	Methylophaga nitratireducitricrescens M1K	KM192265	
BRgam10	Pseudomonadaceae	0	0	0	1	6	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	12	Susp.	Pseudomonas kunmingensis OA009	KY622678	
KTGgam11	Pseudomonadaceae	2	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	4	Source	Pseudomonas pelagia Arc-5-528	KJ630905	
BRgam12	Pseudoalteromonadaceae	0	0	0	0	0	0	0	0	0	0	0	0	2	9	1	0	0	0	0	0	0	12	Susp.	Pseudoalteromonas gangetensis WAB2138	MH169337	
KTGgam13	Pseudoalteromonadaceae	5	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	6	Source	Pseudoalteromonas carrageenovora IAM 12662	LT965929	
KTGgam14	Oceanospirillaceae	47	18	0	4	1	1	0	1	0	0	6	0	1	1	0	1	9	1	0	1	0	97	Source	Marinobacterium georgiense NBRC 102196	AB681723	
BRgam15	Oceanospirillaceae	0	0	0	0	0	0	2	10	23	9	3	2	0	0	0	0	0	1	0	0	0	60	Cathode	Neptunomonas japonica DL5-2-2	KF146511	
BRgam16	Sedimenticolaceae	0	0	0	0	0	0	0	1	0	0	7	0	0	0	0	0	0	0	0	0	0	9	Susp.	Sedimenticola thiotaurini SIP-G1	NR_136790	
BRgam17	Shewanellaceae	0	0	0	0	1	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	4	Susp.	Shewanella algae Hiro-1	LC339942	
BRgam18	Alcanivoracaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	4	0	8	Susp.	Alcanivorax dieselolei B	KP337609	
Epsilonproteobacteria																											
BRReps1	Campylobacteraceae	0	0	0	1	0	1	0	1	1	0	0	0	1	3	2	0	2	2	0	0	0	15	Cathode	Arcobacter haliotis	AB542077	
KTGeps2	Campylobacteraceae	0	0	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	Source	Arcobacter marinus CL-S1	NR_116342	
Betaproteobacteria																											
BRbet1	Rhodocyclaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	5	1	7	Susp.	Azoarcus taiwanensis NSC3	GQ389714	
Actinobacteria																											
BRact1	Coriobacteriaceae	0	0	0	0	0	0	0	0	0	0	0	0	5	0	0	1	0	3	2	0	2	22	Anode	Uncultured bacterium clone OTU136	KU998399	

Phylum-Class		Source			A1		A2		A4		A5		B1		B2		B3		B4		B5		SUM (%)	Location	%match	Closest relative	Accession number	
OTU ID	Family	KTG1	KTG2	KTG3	Anode	Susp.	Cathode	Anode	Susp.	Cathode	Anode	Susp.	Cathode	Anode	Susp.	Cathode	Anode	Susp.	Cathode	Anode	Susp.	Cathode						
Bacteroidetes																												
BRbac1	Lentimicrobiaceae	0	0	0	0	0	0	0	0	0	0	0	0	4	22	4	6	0	0	0	0	0	46	Cathode	100	Lentimicrobium saccharophilum 1S-PR-S3-s2	MG264261	
BRbac9	Lentimicrobiaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	26	4	34	Susp.	95	Lentimicrobium saccharophilum 1S-PR-S3-s2	MG264261
KTGbac2	Flavobacteriaceae	0	5	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	5	Source	99	Winogradskyella epiphytica KMM 3906	NR_025803	
BRbac3	Flavobacteriaceae	0	0	0	0	1	5	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	6	Cathode	100	Muricauda lutimaris 003-Na2	MG456881	
KTGbac4	Flavobacteriaceae	4	10	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	14	Source	98	Planktosalinus lacus X14M-14	NR_149250	
KTGbac5	Bacteroidetes VC2.1 Bac22	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	Source	100	Uncultured bacterium clone BIOP079_s7_OTU16	MF320709	
BRbac6	Draconibacteriaceae	0	0	0	1	1	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	6	Cathode	100	Draconibacterium sediminis MCCC 1A00734	NR_136493	
KTGbac7	Cryomorphaceae	2	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	5	Source	98	Cryomorphaceae bacterium MOLA1068	KX818049	
BRbac8	Porphyromonadaceae	0	0	0	0	0	0	0	0	0	0	9	14	6	0	0	0	0	0	0	0	0	29	Cathode	98	Petrimonas mucosa ING2-E5A	NR_148808	
Cloacimonetes																												
BRclo1	class MSBL8	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	5	4	5	7	3	7	55	Biofilm	96	Uncultured bacterium clone Nit5Au0613_740	FJ628334	
BRclo2	class LK-44f	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	0	1	0	0	0	3	Anode	100	Uncultured bacterium clone SS WC_20	FJ973590	
Firmicutes																												
KTGfir1	Eubacteriaceae	0	0	16	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	16	Source	100	Acetobacterium wieringae DP9	HQ384240	
BRfir2	Clostridiaceae 4	0	0	0	0	0	0	0	0	0	0	0	0	1	10	5	0	0	0	0	0	0	16	Susp.	97	Wukongibacter baidiensis DY30321	NR_152683	
KTGfir3	Clostridiaceae 2	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	Source	99	Tindallia texcoconensis IMP-300	NR_043664	
KTGfir4	Family XII	3	0	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	6	Source	97	Clostridia bacterium TSAR14	KC854383	
Planctomycetes																												
BRpla1	Class Phycisphaerae	0	0	0	0	0	0	15	0	10	3	0	1	0	0	0	0	0	0	0	0	0	30	Anode	95	Uncultured planctomycete MERTZ_21CM_213	AF424493	
Spirochaetae																												
KTGspi1	Spirochaetaceae	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	Source	100	Uncultured bacterium clone JCC_RecOTU_01	KX550146	
BRspi2	Spirochaetaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	2	Cathode	94	Spirochaetaceae bacterium Tc33_10	AB518746	
BRspi3	Spirochaetaceae	0	0	0	0	0	0	4	2	3	2	0	1	2	0	0	0	0	0	0	0	0	15	Biofilm	99	Pleomorphochaeta caudata SEBR 4223	KU714929	
BRspi4	Spirochaetaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	0	3	1	0	1	7	Biofilm	95	Sediminispirochaeta smaragdinae SEBR 4228	NR_027585	
Synergistetes																												
BRsyn1	Synergistaceae	0	0	0	0	0	0	12	5	11	17	8	8	25	12	11	1	2	3	3	3	3	207	Biofilm	92	Thermovirga lienii DSM 17291	NR_043522	
BRsyn2	Synergistaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	7	4	24	Biofilm	91	Thermovirga lienii LABEM006	KJ576629	
BRsyn3	Synergistaceae	0	0	0	0	0	0	0	0	0	5	6	3	0	1	0	0	0	0	0	0	0	15	Cathode	90	Thermovirga lienii LABEM006	KJ576629	
BRsyn4	Synergistaceae	0	0	0	0	0	0	6	1	5	0	0	0	0	0	0	1	0	1	0	0	0	15	Biofilm	90	Thermovirga lienii LABEM006	KJ576629	
BRsyn5	Synergistaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	4	Anode	94	Thermovirga lienii DSM 17291	NR_043522		
Chloroflexi																												
BRchl1	Anaerolineaceae	0	0	0	0	0	0	0	0	0	2	0	1	0	0	0	0	0	0	0	0	0	8	Anode	94	Pelolinea submarina MO-CFX1	NR_133813	
BRchl2	Anaerolineaceae	0	0	0	0	0	0	0	0	0	0	0	0	4	1	2	3	0	5	2	0	2	24	Anode	88	Anaerobic bacterium MO-CFX2	AB598278	
BRchl3	Anaerolineaceae	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	1	0	36	Biofilm	97	Brevefilum fermentans CAMBI-1	LT859958	
BRchl4	Anaerolineaceae	0	0	0	0	0	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	0	3	Anode	92	Anaerobic bacterium MO-CFX2	AB598278	
BRchl5	Anaerolineaceae	0	0	0	0	0	0	0	0	0	0	0	0	2	0	1	1	0	0	0	0	0	4	Anode	92	Anaerobic bacterium MO-CFX2	AB598278	
Tenericutes																												
BRten1	Acholeplasmataceae	0	0	0	0	0	0	0	0	0	0	0	0	5	5	4	0	0	0	0	0	0	14	Biofilm	100	Tenericutes bacterium MO-XQ	AB598274	
KTGten2	order NB1-n	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	Source	87	Acholeplasma palmae	FO681347	
BRten3	order NB1-n	0	0	0	0	0	0	9	4	14	0	0	0	0	0	0	0	0	0	0	0	0	27	Biofilm	100	Bacterium 4S-PB-a	MG264277	
BRten4	order NB1-n	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	1	1	1	0	1	9	Biofilm	95	Bacterium 4S-PB-a	MG264277	
Thermotogae																												
BRthr1	Thermotogaceae	0	0	0	0	0	0	0	0	0	5	1	2	0	0	1	3	4	6	2	1	3	46	Biofilm	99	Mesotoga prima MesG1.Ag.4.2	NR_102952	
others																												
BRmisc1	Hyd24-12	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	0	1	0	0	1	6	Biofilm	87	Caldithrix palaeochoryensis 3151	KU306334	
Euryarchaeota																												
BReur1	Methanobacteriaceae	0	0	0	0	0	0	0	0	0	0	0	0	1	1	7	3	4	7	1	4	6	68	Cathode	100	Methanobacterium sp. Mba6	LC183834	
BReur2	Methanobacteriaceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	2	0	1	1	21	Cathode	100	Methanobacterium formicicum Mb8	JN205059	
BReur3	Methanobacteriaceae	0	0	0	0	0	0	0	0	0	0	1	12	0	0	0	0	0	0	0	0	0	39	Cathode	100	Methanobacterium aarhusense H2-LR	NR_042895	
BReur4	Methanobacteriaceae	0	0	0	0	0	0	0	0	0	0	0	0	4	12	33	5	11	10	1	2	8	94	Cathode	100	Methanobrevibacter arboriphilus	AB065294	
BReur5	Methanocalculaceae	0	0	0	0	0	0	2	24	11	4	11	62	0	0	0	2	8	6	1	4	10	214	Cathode	100	Methanocalculus pumilus MHT-1	NR_028148	
BReur6	Methanosaeataceae	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	14	3	19	133	Biofilm	99	Methanosaeata harundinacea 6Ac	CP003117	