Figure S1

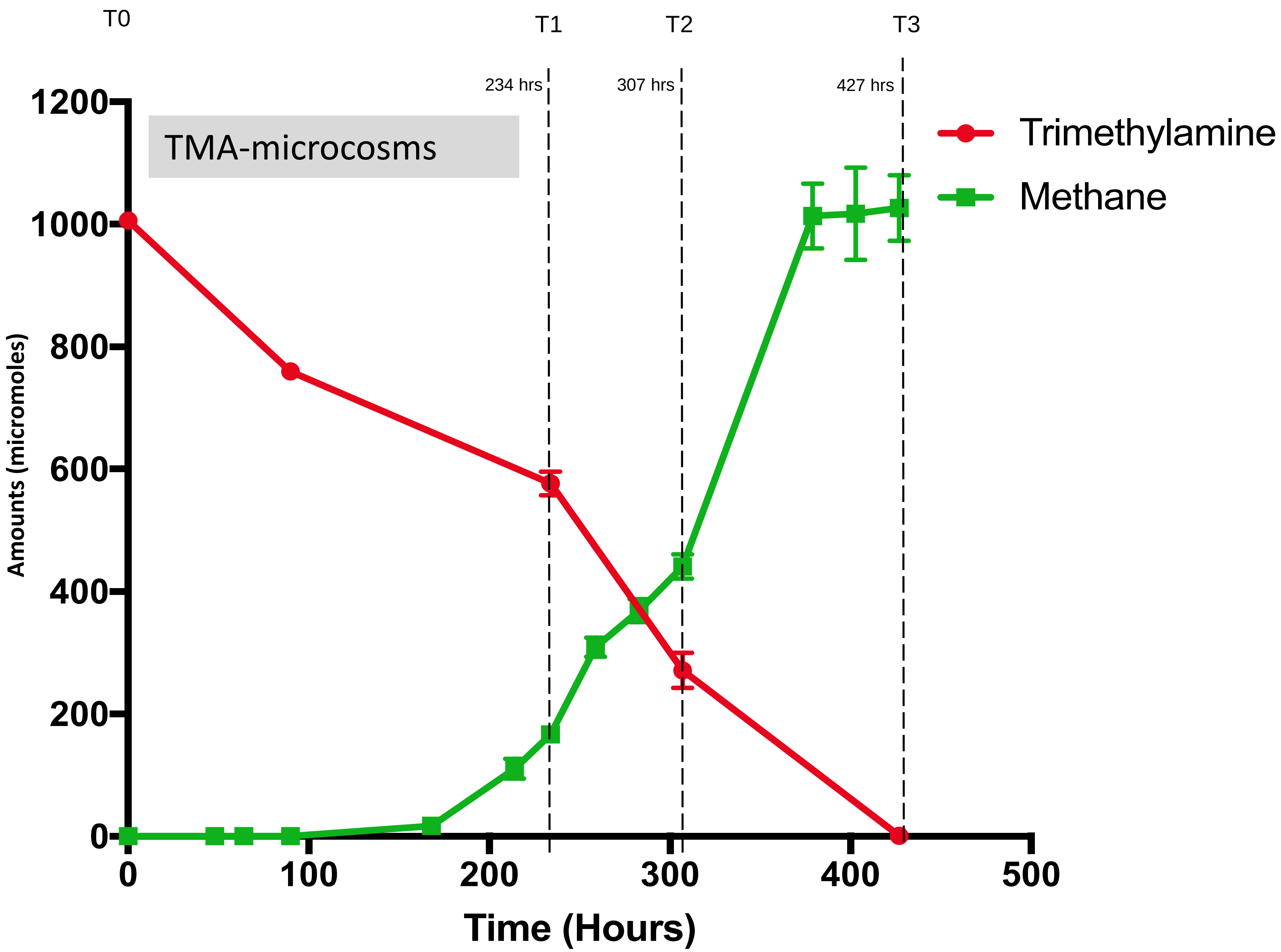

T0
T1
T2
T3
234 hrs
307 hrs
427 hrs
TMA-microcosms
Trimethylamine
Methane
Amounts (micromoles)
1200
1000
800
600
400
200
0
0
100
200
300
400
500
Time (Hours)

Figure S2

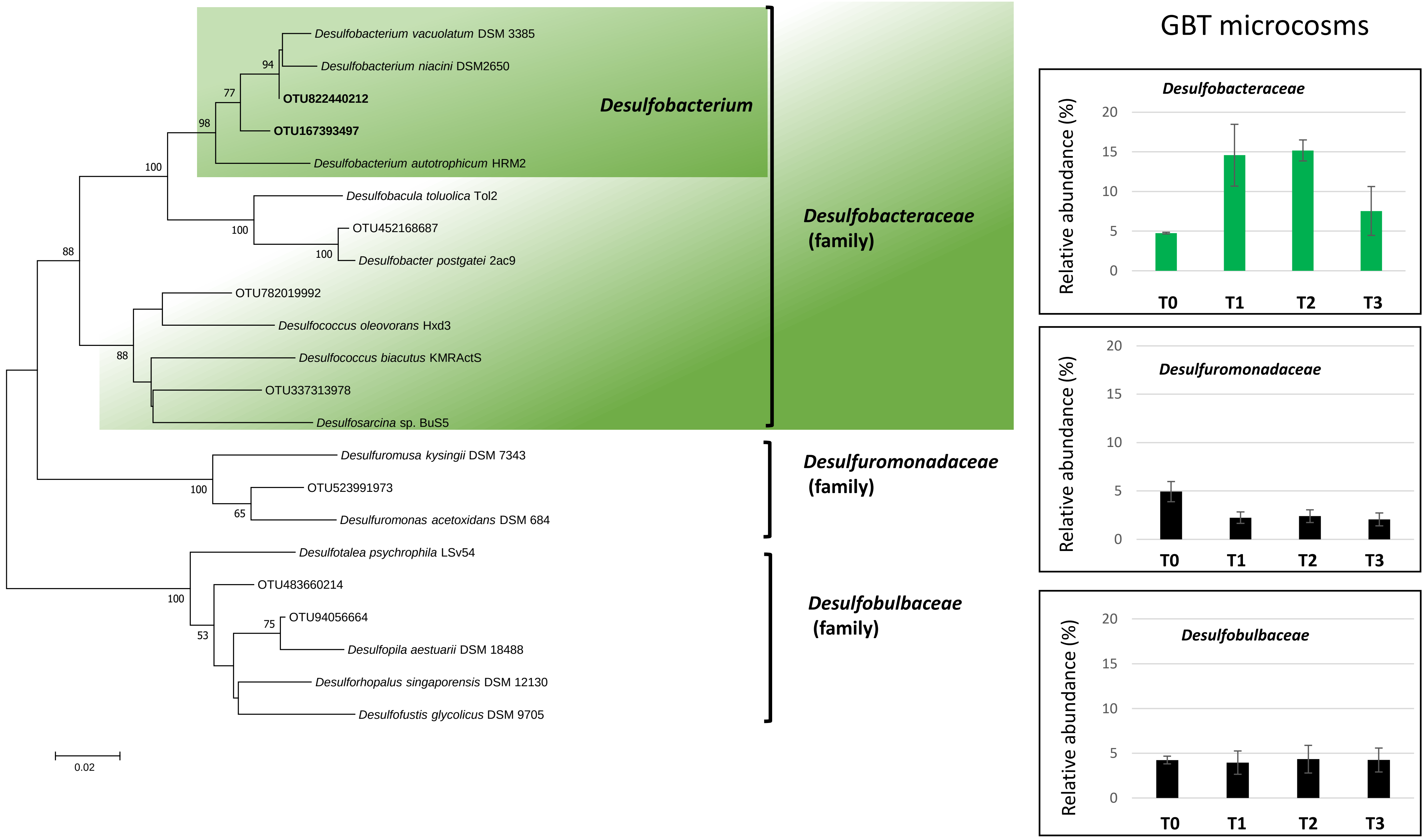
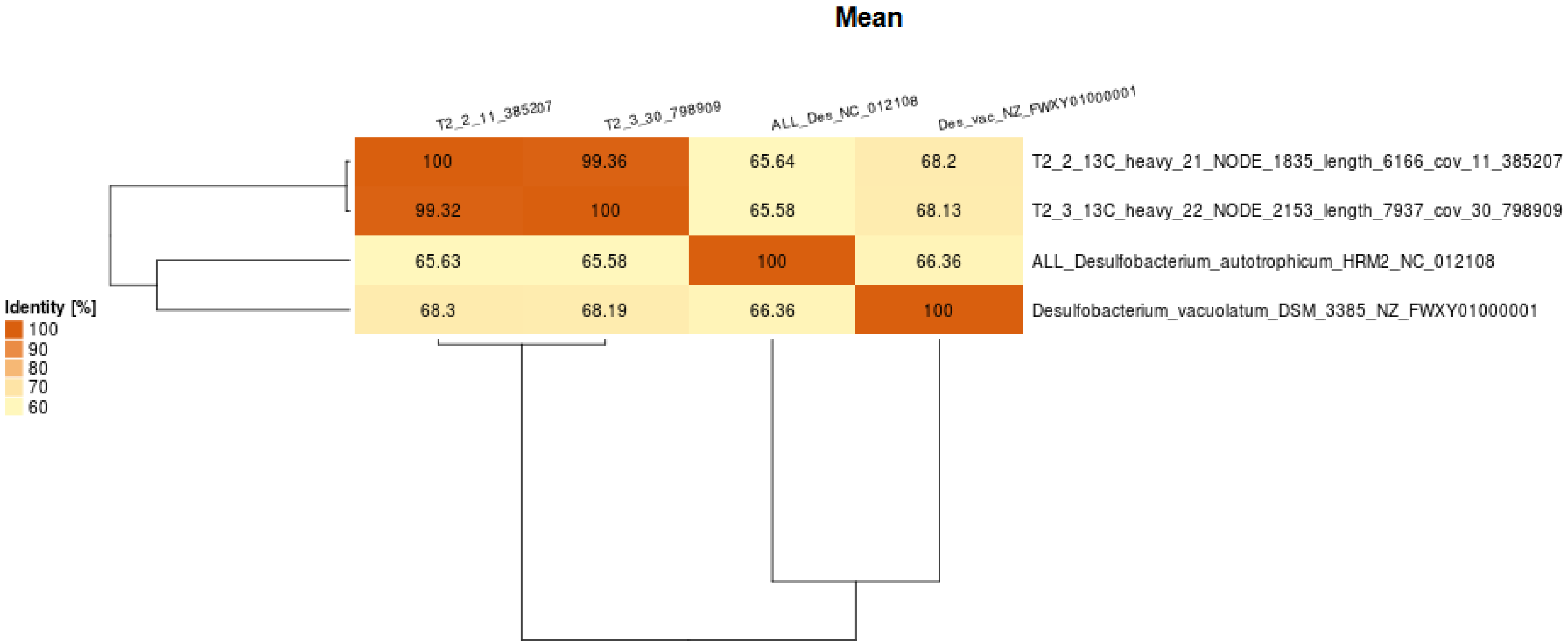

GBT microcosms
Desulfobacterium vacuolatum DSM 3385
Desulfobacterium niacini DSM2650
OTU822440212
OTU167393497
Desulfobacterium autotrophicum HRM2
Desulfobacula toluolica Tol2
OTU452168687
Desulfobacter postgatei 2ac9
OTU782019992
Desulfococcus oleovorans Hxd3
Desulfococcus biacutus KMRActS
OTU337313978
Desulfosarcina sp. BuS5
Desulfuromusa kysingii DSM 7343
OTU523991973
Desulfuromonas acetoxidans DSM 684
Desulfotalea psychrophila LSv54
OTU483660214
OTU94056664
Desulfopila aestuarii DSM 18488
Desulforhopalus singaporensis DSM 12130
Desulfofustis glycolicus DSM 9705
94
77
98
100
100
100
88
88
100
65
100
53
75
0.02
Desulfobacterium
Desulfobacteraceae (family)
Desulfuromonadaceae (family)
Desulfobulbaceae (family)
Desulfobacteraceae
Desulfuromonadaceae
Desulfobulbaceae
Relative abundance (%)
0
5
10
15
20
T0
T1
T2
T3

Figure S3

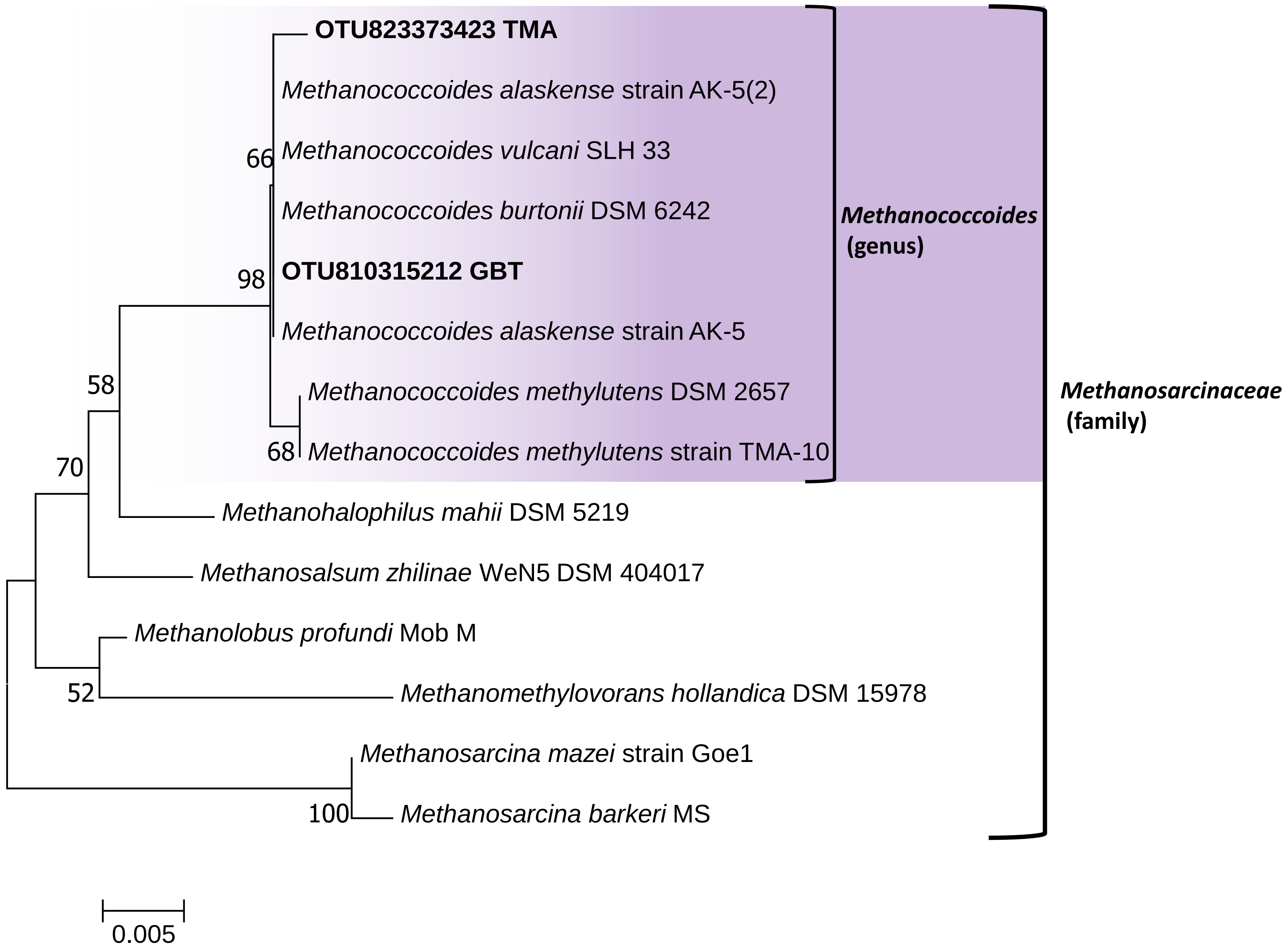
Genome characteristics of two *Desulfobacterium*-related population genomes extracted from metagenomes and two *Desulfobacterium* genomes

Organism	IMG genome ID	Genome size (Number of total bases)	Gene count (Number of total genes)	CDS count (Number of CDS genes)	rRNA count (Number of rRNA genes)	Protein coding genes with function prediction	Protein coding genes without function prediction
13C Heavy 2 Bin 21	-	6,489,886			26	3443	2156
13C Heavy 3 Bin 22	-	6,624,919			34	3680	2217
D. autotrophicum DMS 3383	643692021	5,657,782	5012	4943	69	3552	1391
D. vacuolatum DSM 3385	2681813565	5,035,799	4183	4101	82	3302	799



	T2_2_11_385207	T2_3_30_798909	ALL_Des_NC_012108	Des_vac_NZ_FWXY01000001
T2_2_13C_heavy_21_NODE_1835_length_6166_cov_11_385207	100	99.36	65.64	68.2
T2_3_13C_heavy_22_NODE_2153_length_7937_cov_30_798909	99.32	100	65.58	68.13
ALL_Desulfobacterium_autotrophicum_HRM2_NC_012108	65.63	65.58	100	66.36
Desulfobacterium_vacuolatum_DSM_3385_NZ_FWXY01000001	68.3	68.19	66.36	100

Figure S4


OTU823373423 TMA
Methanococcoides alaskense strain AK-5(2)
Methanococcoides vulcani SLH 33
Methanococcoides burtonii DSM 6242
OTU810315212 GBT
Methanococcoides alaskense strain AK-5
Methanococcoides methylutens DSM 2657
Methanococcoides methylutens strain TMA-10
Methanohalophilus mahii DSM 5219
Methanosalsum zhilinae WeN5 DSM 404017
Methanolobus profundi Mob M
Methanomethylovorans hollandica DSM 15978
Methanosarcina mazei strain Goe1
Methanosarcina barkeri MS
66
98
58
70
68
52
100
Methanococcoides (genus)
Methanosarcinaceae (family)
0.005

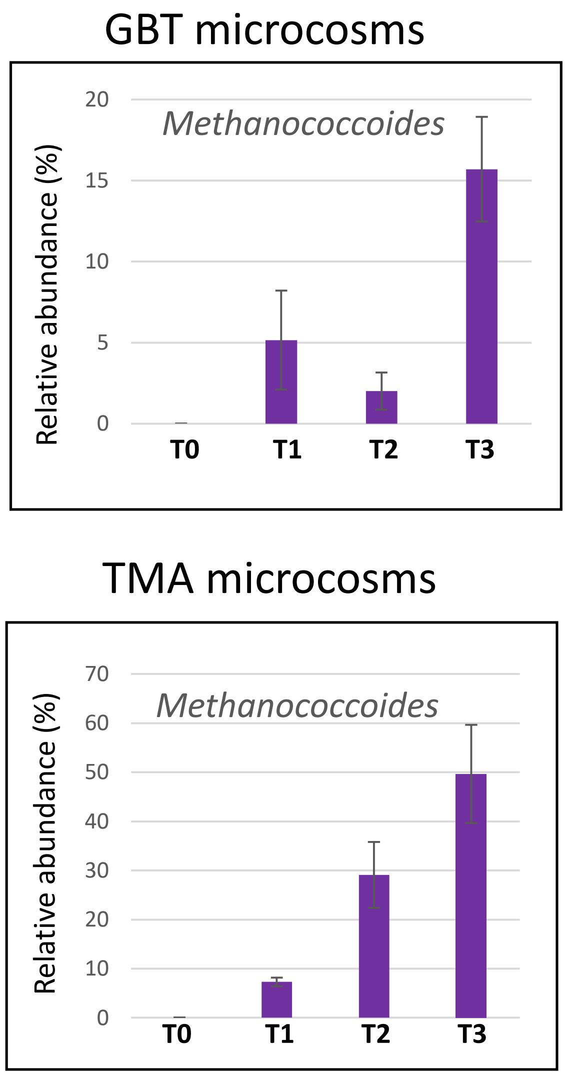
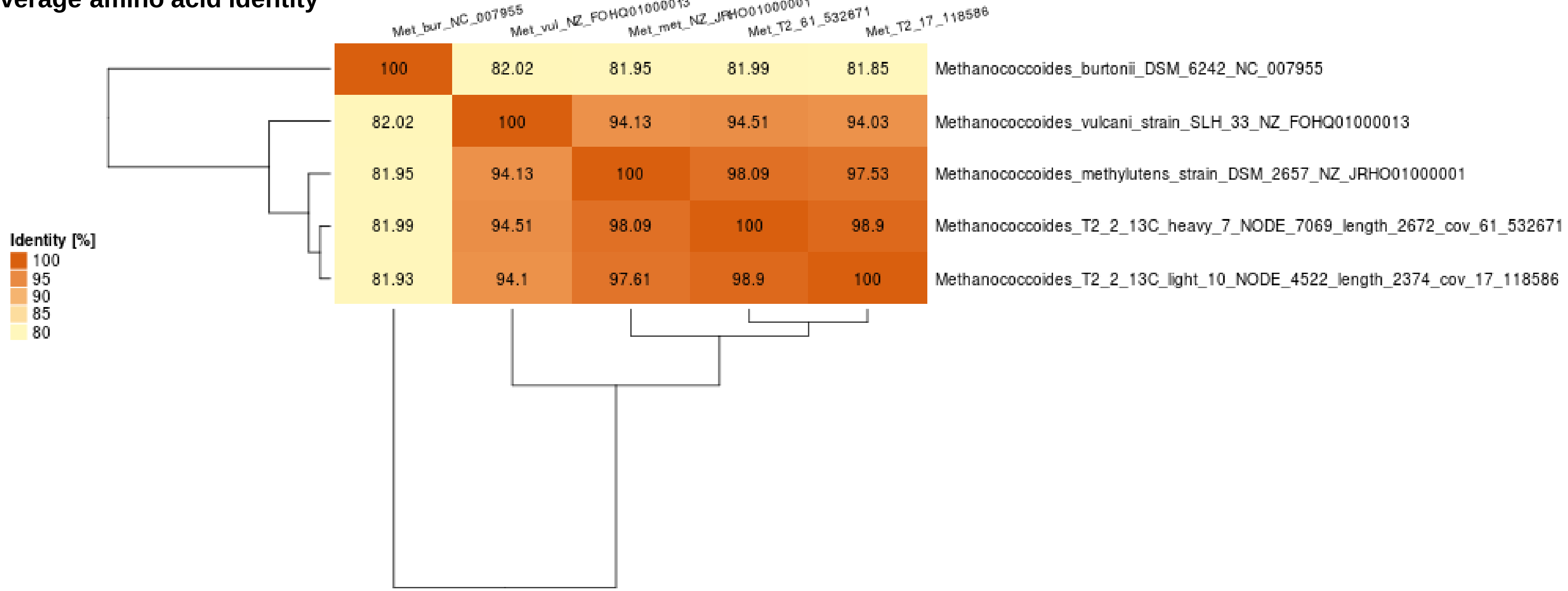

GBT microcosms
Methanococcoides
Relative abundance (%)
0
5
10
15
20
T0
T1
T2
T3
TMA microcosms
Methanococcoides
Relative abundance (%)
0
10
20
30
40
50
60
70
T0
T1
T2
T3

Figure S5

Genome characteristics of two *Methanococcoides*-related population genomes extracted from metagenomes and three *Methanococcoides* genomes

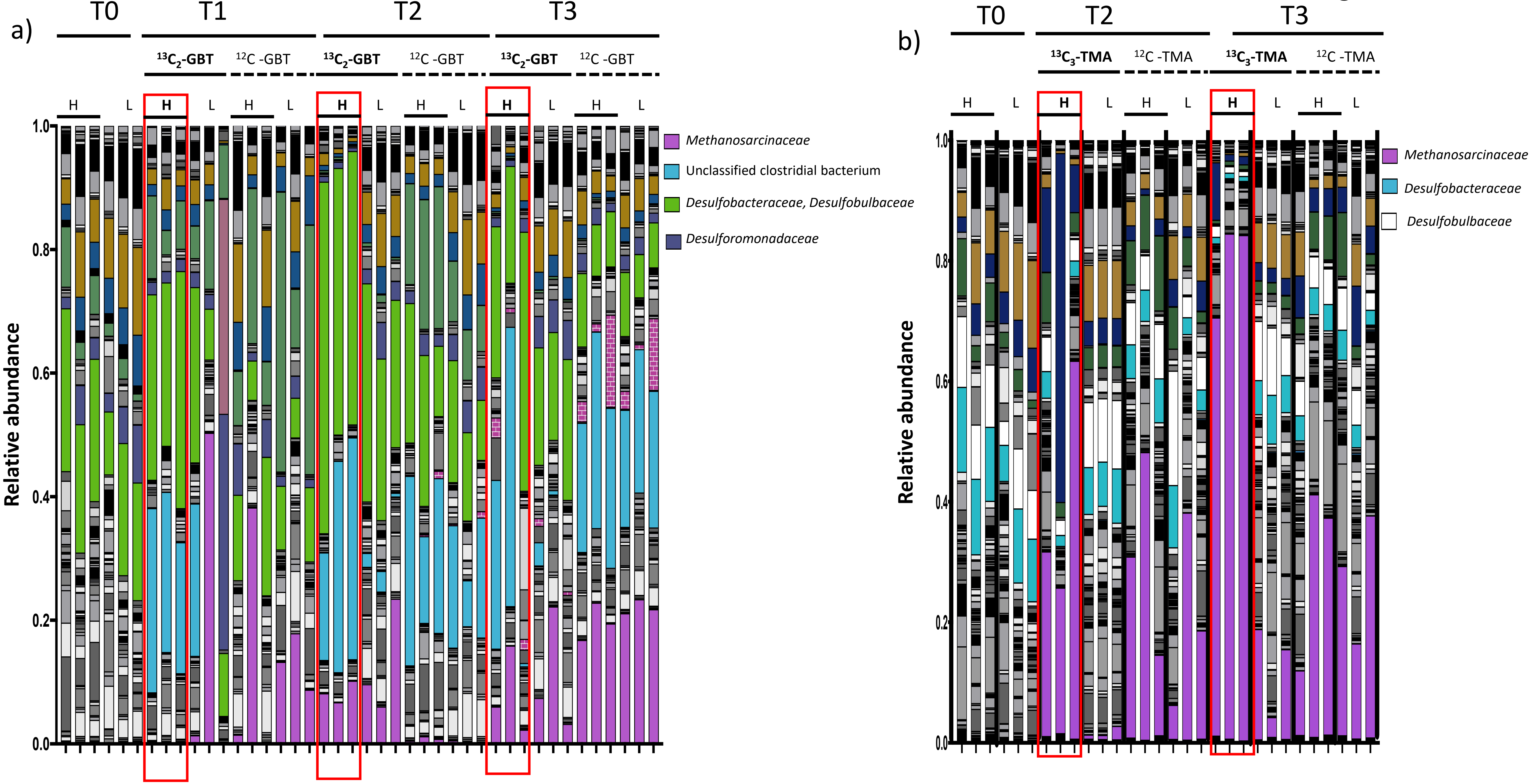
Organism	IMG genome ID	Genome size (Number of total bases)	Gene count (Number of total genes)	CDS count (Number of CDS genes)	rRNA count (Number of rRNA genes)	Protein coding genes with function prediction	Protein coding genes without function prediction
13C Heavy 2 Bin 7	-	1,564,734			24	1,105	378
13C Light 2 Bin 10	-	924,130			14	725	186
M. vulcani SLH 33	2642422537	2,314,846	2,357	2,290	67	1,712	578
M. burtonii DSM 6242	637000161	2,575,032	2,506	2,431	75	1,566	865
M. methylutens DSM 2657	2630968972	2,508,511	2,527	2,462	10	1,805	657

Average amino acid identity



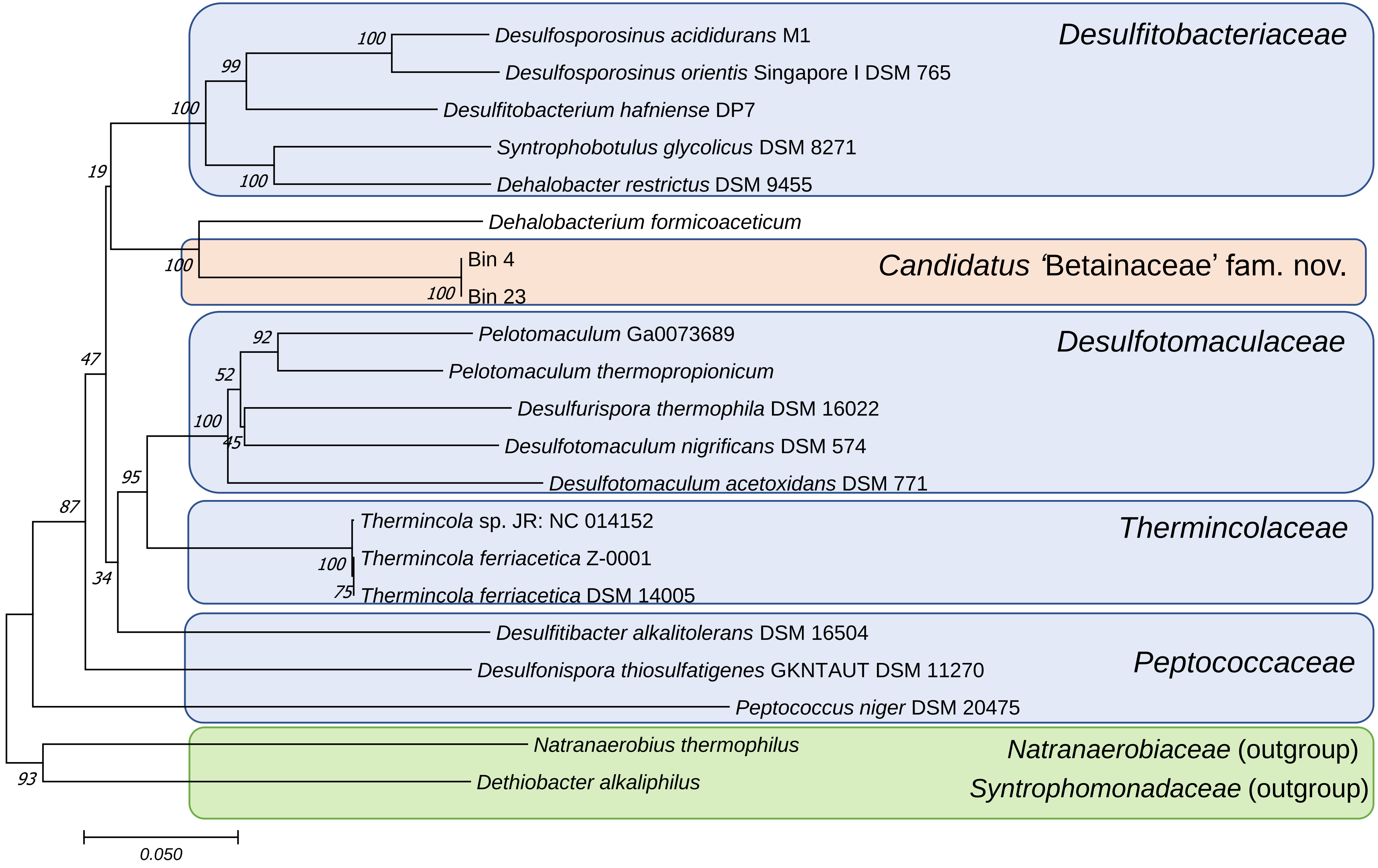
Met_bur_NC_007955	Met_vul_NZ_FOHQ01000013	Met_met_NZ_JRHO01000001	Met_T2_61_532671	Met_T2_17_118586	
100	82.02	81.95	81.99	81.85	Methanococcoides_burtonii_DSM_6242_NC_007955
82.02	100	94.13	94.51	94.03	Methanococcoides_vulcani_strain_SLH_33_NZ_FOHQ01000013
81.95	94.13	100	98.09	97.53	Methanococcoides_methylutens_strain_DSM_2657_NZ_JRHO01000001
81.99	94.51	98.09	100	98.9	Methanococcoides_T2_2_13C_heavy_7_NODE_7069_length_2672_cov_61_532671
81.93	94.1	97.61	98.9	100	Methanococcoides_T2_2_13C_light_10_NODE_4522_length_2374_cov_17_118586

Figure S6



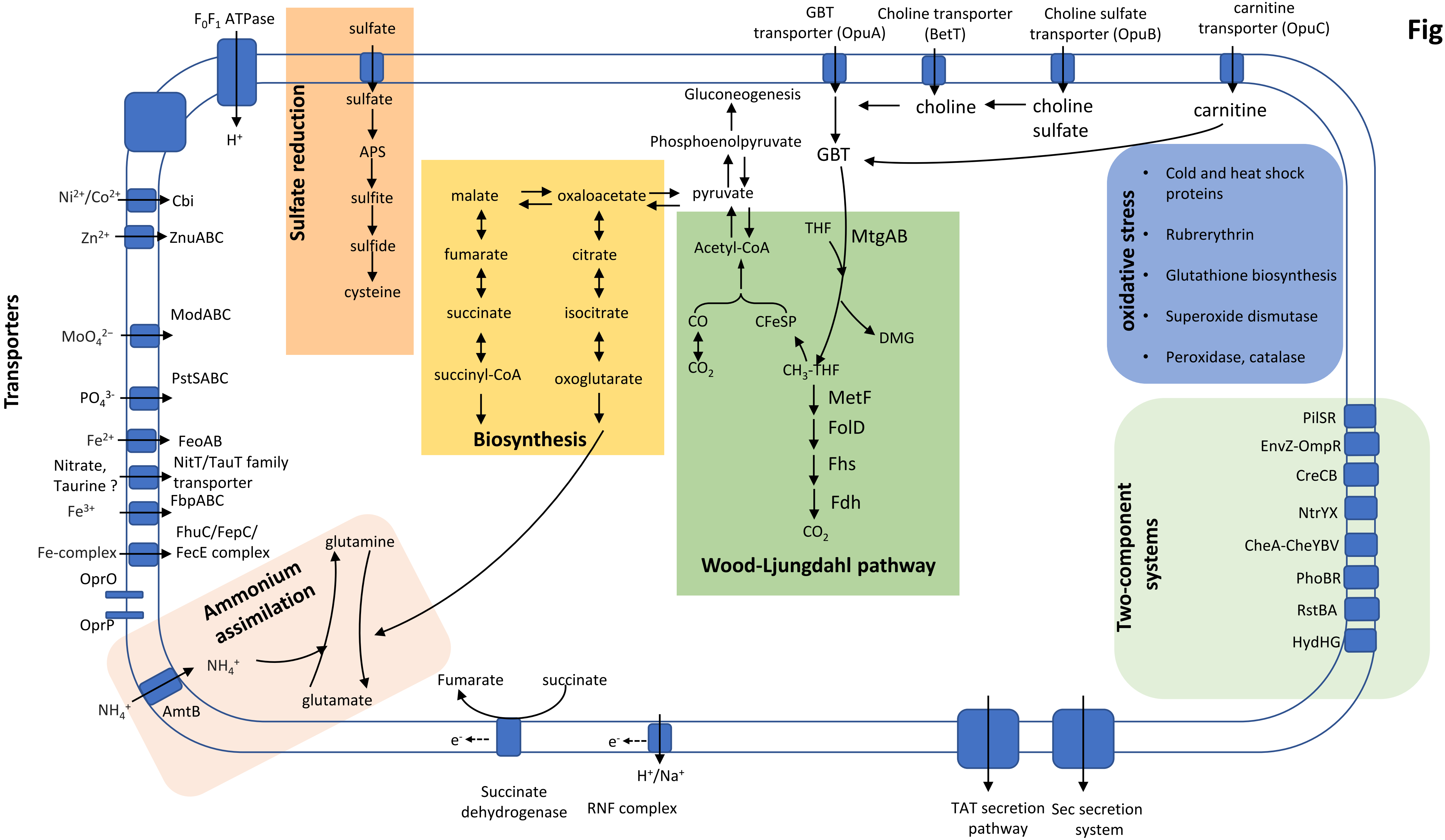
a)
T0
T1
T2
T3
$^{13}C_2$-GBT
^{12}C -GBT
H
L
Relative abundance
1.0
0.8
0.6
0.4
0.2
0.0
Methanosarcinaceae
Unclassified clostridial bacterium
Desulfobacteraceae, Desulfobulbaceae
Desulforomonadaceae
b)
T0
T2
T3
$^{13}C_3$-TMA
^{12}C -TMA
H
L
Relative abundance
1.0
0.8
0.6
0.4
0.2
0.0
Methanosarcinaceae
Desulfobacteraceae
Desulfobulbaceae

Figure S7



Desulfitobacteriaceae
100
Desulfosporosinus acididurans M1
99
Desulfosporosinus orientis Singapore I DSM 765
100
Desulfitobacterium hafniense DP7
19
Syntrophobotulus glycolicus DSM 8271
100
Dehalobacter restrictus DSM 9455
Dehalobacterium formicoaceticum
100
Candidatus 'Betainaceae' fam. nov.
Bin 4
100
Bin 23
Desulfotomaculaceae
92
Pelotomaculum Ga0073689
47
52
Pelotomaculum thermopropionicum
Desulfurispora thermophila DSM 16022
100
45
Desulfotomaculum nigrificans DSM 574
95
Desulfotomaculum acetoxidans DSM 771
87
Thermincolaceae
Thermincola sp. JR: NC 014152
100
Thermincola ferriacetica Z-0001
34
75
Thermincola ferriacetica DSM 14005
Desulfitibacter alkalitolerans DSM 16504
Peptococcaceae
Desulfonispora thiosulfatigenes GKNTAUT DSM 11270
Peptococcus niger DSM 20475
Natranaerobius thermophilus
Natranaerobiaceae (outgroup)
93
Dethiobacter alkaliphilus
Syntrophomonadaceae (outgroup)
0.050

Figure S8


F_0F_1 ATPase
H^+
Sulfate reduction
sulfate
sulfate
APS
sulfite
sulfide
cysteine
GBT transporter (OpuA)
Choline transporter (BetT)
Choline sulfate transporter (OpuB)
carnitine transporter (OpuC)
Gluconeogenesis
choline
choline sulfate
carnitine
Phosphoenolpyruvate
GBT
pyruvate
malate
oxaloacetate
fumarate
citrate
succinate
isocitrate
succinyl-CoA
oxoglutarate
Biosynthesis
Acetyl-CoA
THF
MtgAB
CO
CFeSP
CO_2
CH_3-THF
DMG
MetF
FolD
Fhs
Fdh
CO_2
Wood-Ljungdahl pathway
oxidative stress
Cold and heat shock proteins
Rubrerythrin
Glutathione biosynthesis
Superoxide dismutase
Peroxidase, catalase
Transporters
Ni^{2+}/Co^{2+}
Cbi
Zn^{2+}
ZnuABC
ModABC
MoO_4^{2-}
PstSABC
PO_4^{3-}
Fe^{2+}
FeoAB
Nitrate, Taurine ?
NitT/TauT family transporter
FbpABC
Fe^{3+}
FhuC/FepC/FecE complex
Fe-complex
OprO
OprP
Ammonium assimilation
glutamine
NH_4^+
glutamate
NH_4^+
AmtB
Fumarate
succinate
e^-
e^-
H^+/Na^+
Succinate dehydrogenase
RNF complex
TAT secretion pathway
Sec secretion system
Two-component systems
PilSR
EnvZ-OmpR
CreCB
NtrYX
CheA-CheYBV
PhoBR
RstBA
HydHG