Supplementary material

Figures:

Figure S1. Monthly methane anomaly in the surface layer at 0, 5, 10, and 20 m. Positive and negative anomalies are shown in red and blue, respectively. It was at a fixed station (ST18) on the central Chile upwelling platform during 2018 and 2021.

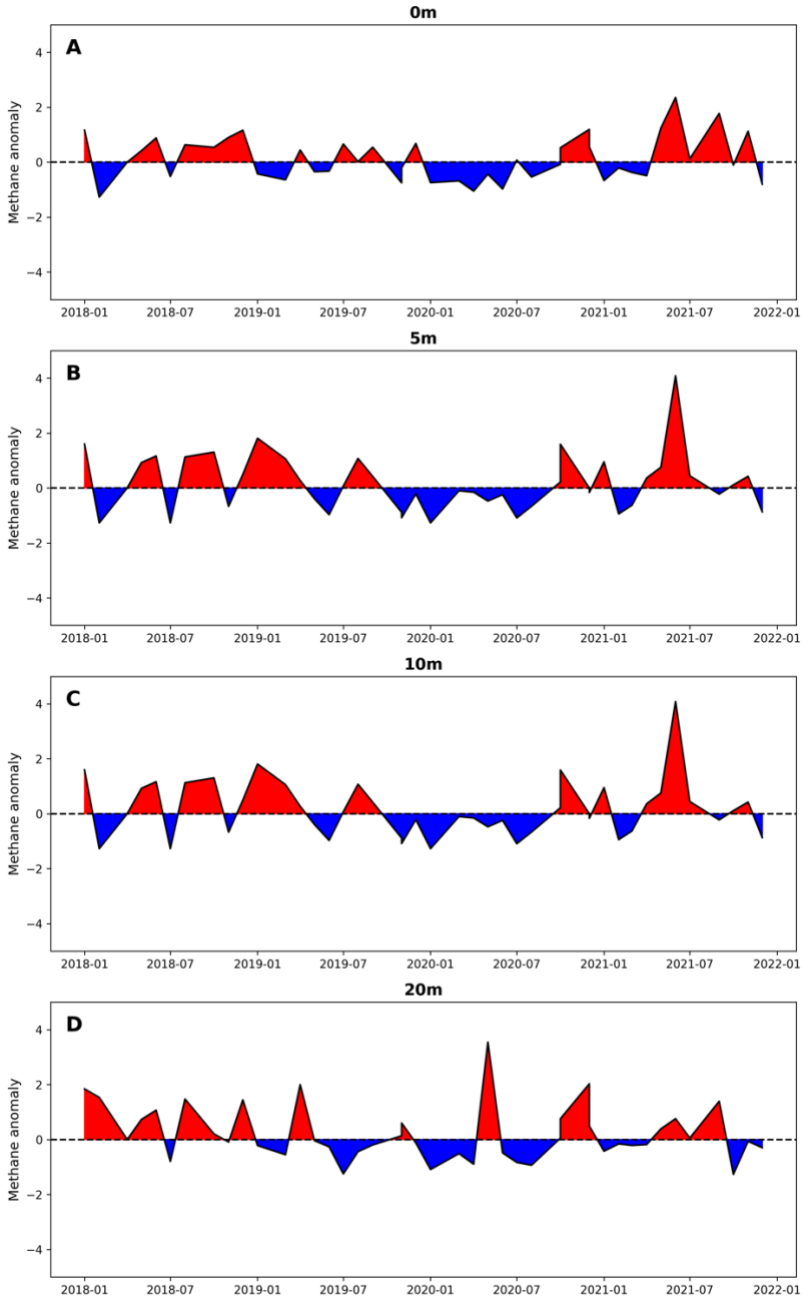


Figure S2. Spearman correlations matrix between biochemical variables at the surface layer (20m). Values on the red and blue color scale represent positive and negative values of the Spearman correlation (rho). Variables are: T: temperature, S: salinity, DO: dissolved oxygen; NO_3^- ; nitrate; PO_4^{3-}: phosphate; $Si(OH)_4$: silicic acid; Chl-a, chlorophyll-a; CH_4: methane; N:P ratio, Si:N ratio and DOC: dissolved oxygen carbon. * represents a significant correlation of 0.05.

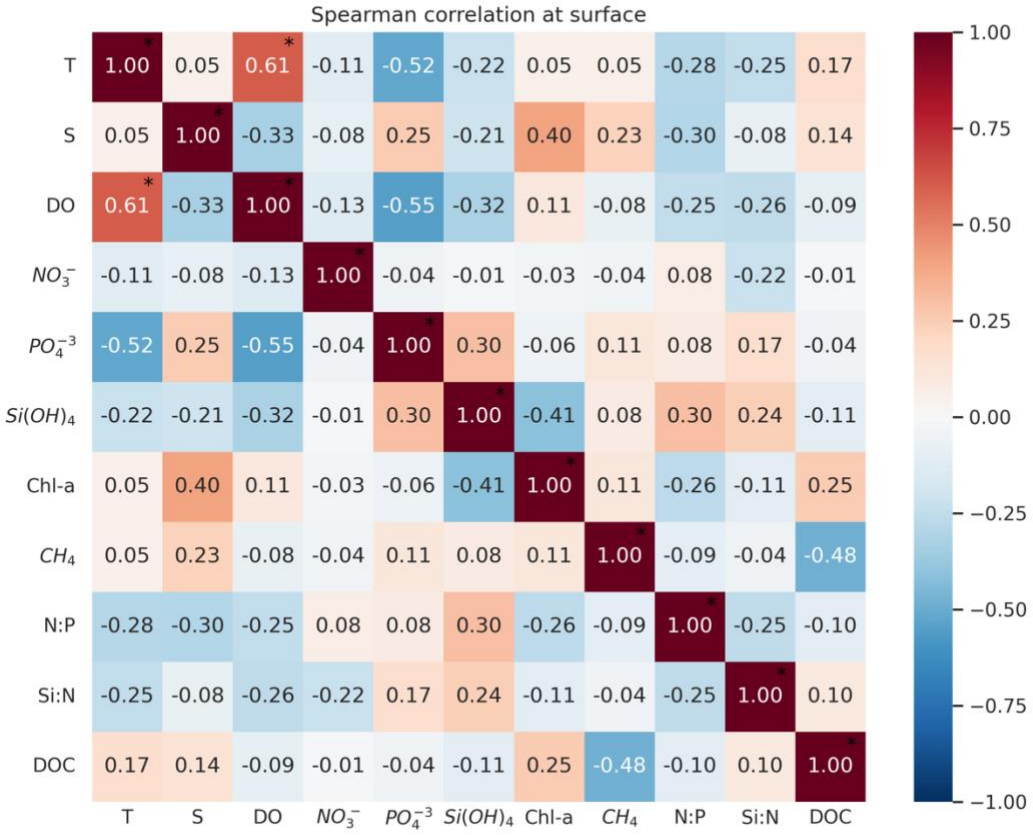
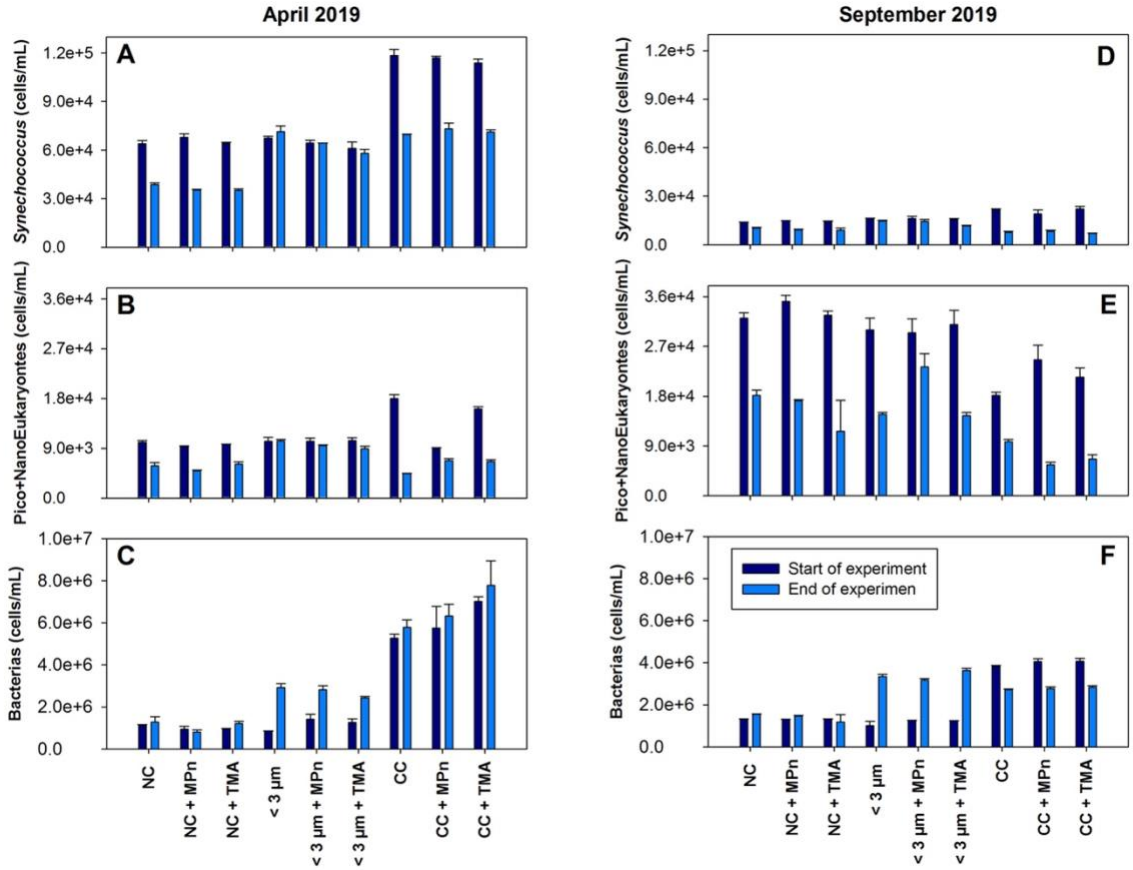


Figure S3. Abundance of *Synechococcus sp.* (cell mL^{-1}), pico + nano eukaryotes (cell mL^{-1}), and bacteria (cell mL^{-1}) at the beginning (dark blue) and at the end (light blue) of the microcosm experiments with the addition of methylated substrates (MPn: methyl phosphonic acid and TMA: trimethylamine) performed with three planktonic communities (NC: natural planktonic community; <3 µm: picoplankton and CC: picoplankton concentrate) under oxygenated conditions in April (A, B, and C) and September 2019 (D, E, and F).



Tables:

Table S1. Initial concentrations of biogeochemical variables in methane cycling experiments with the following treatments: NC: natural community; <3 μm: picoplankton and <0.2 μm: femtoplankton; and with the addition of methylated substrate (MPn: methyl phosphonic acid and TMA: trimethylamine) conducted in December 2018, January, March, and May 2019.

Month	Treatments (μm)	CH_4 (nM)	OD (μM)	Chl-a (mg m^{-3})	DOC (μM)	NO_3^- (μM)	NO_2^- (μM)	PO_4^{3-} (μM)	$Si(OH)_4$ (μM)
Dec	CN	2.21	274.67	43.81	101.12	1.55	0.14	0.77	0.30
	<3	6.98	273.32	6.50	127.10	1.31	0.13	0.66	0.07
	<0.2	3.88	279.55	-	124.26	2.29	0.14	0.79	0.47
Jan	CN	13.77	185.48	15.70	158.82	16.18	0.33	1.62	4.21
	<3	5.01	235.28	2.39	98.36	15.86	0.35	1.69	3.94
	<0.2	3.46	259.79	-	123.60	17.46	0.36	1.91	5.53
Mar	<3 Ctrl	8.65	187.81	1.39	73.94	6.61	0.60	1.54	5.29
	<3+MPn	22.74	218.23	0.44	88.73	5.92	0.56	1.53	4.56
May	<3 Ctrl	17.41	275.27	0.46	166.68	9.71	0.30	1.15	3.15
	<3+MPn	16.25	268.94	0.41	123.06	13.45	0.40	1.25	4.24
	<3+TMA	18.15	268.74	0.43	149.62	13.69	0.43	1.37	4.49

Table S2. Net rates of methane cycling (net accumulation and/or consumption) under different treatments: NC: natural plankton community and <3 μm: picoplankton; and with the addition of methylated substrates (MPn: methyl phosphonic acid and TMA: trimethylamine) in December 2018, January, March, and May 2019.

Month	Treatments (μm)	Total	Light	Dark
Dec	CN	0.10 ± 0.04	0.10 ± 0.06	0.08 ± 0.27
	<3	0.3 ± 0.07	0.11 ± 0.11	0.59 ± 0.13 (*)
Jan	CN	-0.41 ± 0.16	-0.28 ± 0.41	-0.04 ± 0.1
	<3	-0.46 ± 0.17	-0.41 ± 0.5	-0.28 ± 0.24
Mar	<3 Ctrl	0.16 ± 0.11	-0.51 ± 0.23	0.28 ± 0.10 (*)
	<3+MPn	0.05 ± 0.29	-0.99 ± 0.09	-0.71 ± 0.18
May	<3 Ctrl	-0.14 ± 0.09	-0.25 ± 0.12	-0.13 ± 0.33
	<3+MPn	0.17 ± 0.22	0.19 ± 0.33	1.02 ± 0.53 (*)
	<3+TMA	-0.01 ± 0.16	0.21 ± 0.60 (*)	0.25 ± 0.08 (*)

Table S3. Initial concentrations of biogeochemical variables in long-term microcosm experiment with the addition of methylated substrates (MPn: methyl phosphonic acid and TMA: trimethylamine) conducted over three planktonic communities (NC: natural plankton community; <3 μm: picoplankton and CC: picoplankton concentrate) in April 2019 and September 2019.

Month	Treatments (μm)	CH_4 (nM)	OD (μM)	Chl-a (mg m^{-3})	DOC (μM)	NO_3^- (μM)	NO_2^- (μM)	PO_4^{3-} (μM)	$Si(OH)_4$ (μM)
Apr	NC Ctrl	25.13	223.35	8.48	97.27	15.35	0.74	1.95	3.72
	NC + MPn	25.13	211.44	8.47	90.76	14.40	0.71	1.93	4.04
	NC TMA	25.13	202.06	7.36	94.81	13.16	0.62	1.75	3.15
	<3 Ctrl	25.09	209.11	0.39	115.27	13.32	0.66	1.74	3.84
	<3 + MPn	25.06	217.01	0.16	117.97	13.66	0.65	1.73	3.91
	<3 + TMA	25.05	220.72	0.88	118.02	13.69	0.73	1.93	3.69
	CC Ctrl	23.71	273.65	1.76	118.98	14.49	0.43	1.84	4.69
	CC + MPn	24.37	264.15	1.19	156.74	11.76	0.35	1.51	3.90
	CC + TMA	21.05	270.68	1.64	142.50	10.76	0.37	1.47	3.51
Sep	NC Ctrl	21.13	210.03	1.31	162.65	12.44	0.23	2.08	12.09
	NC + MPn	20.43	175.84	1.16	100.93	14.95	0.24	1.68	11.35
	NC TMA	20.33	189.53	1.02	179.15	11.92	0.23	1.73	9.12
	<3 Ctrl	20.08	201.42	0.39	151.80	13.81	0.23	1.85	8.69
	<3 + MPn	20.33	209.76	0.36	102.51	12.89	0.23	1.97	8.63
	<3 + TMA	20.73	192.07	0.35	104.45	12.27	0.15	1.56	8.22
	CC Ctrl	-	194.99	0.85	107.82	13.51	0.11	1.84	9.53
	CC + MPn	-	178.55	0.80	122.47	13.10	0.07	1.77	11.36
	CC + TMA	-	155.93	0.83	174.24	13.59	0.09	1.48	7.91

Table S4. Net rates of methane cycling (net accumulation and/or consumption) under different treatments: NC: natural plankton community; <3 μm: picoplankton and CC: picoplankton concentrate; and the addition of methylated substrates (MPn: methyl phosphonic acid and TMA: trimethylamine), conducted in April and September 2019.

Treatments (μm)	April			September		
	Total	Light	Dark	Total	Light	Dark
NC Ctrl	-0.18 ± 0.04	-0.19 ± 0.06	-0.14 ± 0.01	0.02 ± 0.02	0.03 ± 0.03	-0.02 ± 0.02
NC + MPn	-0.04 ± 0.05	-0.06 ± -0.01	0.07 ± -0.02	-0.03 ± -0.35	-0.02 ± 0.03	-0.11 ± 0.02
NC TMA	-0.23 ± 0.07	-0.24 ± -0.05	-0.13 ± 0.03	-0.04 ± -0.46	-0.03 ± 0.03	-0.11 ± 0.02
<3 Ctrl	-0.02 ± 0.06	-0.03 ± -0.01	0.08 ± -0.01	-0.01 ± -0.09	0.00 ± 0.03	-0.07 ± 0.02
<3 + MPn	-0.17 ± 0.04	-0.18 ± -0.06	-0.12 ± 0.03	0.00 ± -0.03	0.01 ± 0.03	-0.06 ± 0.02
<3 + TMA	0.11 ± 0.03	0.11 ± -3.23	0.14 ± -0.02	0.01 ± 0.18	0.02 ± 0.03	-0.03 ± 0.02
CC Ctrl	-0.07 ± 0.03	-0.08 ± -0.62	-0.02 ± 0.02	-0.02 ± -0.31	-0.03 ± 0.03	0.02 ± 0.01
CC + MPn	0.10 ± 0.02	0.10 ± -0.17	0.12 ± -0.25	-0.07 ± -0.53	-0.09 ± 0.04	0.03 ± 0.01
CC + TMA	0.07 ± 0.02	0.07 ± 0.01	0.06 ± 0.04	0.01 ± 0.08	-0.02 ± 0.03	0.09 ± 0.03