Supplement of Biogeosciences, 21, 4273–4283, 2024
https://doi.org/10.5194/bg-21-4273-2024-supplement

Supplement of

The geothermal gradient shapes microbial diversity and processes in natural-gas-bearing sedimentary aquifers

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Supplementary Tables

Table S1. Depths and water temperatures of the production wells used in this study and their analytical purposes.

Well no.	Sample type	Formations	Strainer depth (average, mbgs)	Water Temp. (°C)	Analysis: Inorganic and organic chemistry	Analysis: Stable sulfur isotope	Analysis: Cell counts	Analysis: Molecular Gene	Analysis: Radiotracer & cultivation
well 1	Formation water	Haizume	554	38	+	+	+	+	+
well 2	Formation water	Haizume & Nishiyama	653	49	+				
well 3	Formation water	Nishiyama	754	51	+		+	+	+
well 4	Formation water	Nishiyama	768	56	+				
well 5	Formation water	Nishiyama	883	54	+				
well 6	Formation water	Nishiyama	798	57	+				
well 7	Formation water	Nishiyama	849	58	+		+	+	+
well 8	Formation water	Nishiyama	845	60	+				
well 9	Formation water	Nishiyama	1049	65	+	+	+	+	+
well 10	Formation water	Nishiyama	1115	73	+	+	+	+	+
well 11	Formation water	Nishiyama	1373	81	+	+	+	+	+
well 12	Formation water	Nishiyama	1374	82	+				
wells 13*	Formation water & oil	Nishiyama & Shiiya	1513	67	+	+			
wells 14*	oil	Shiiya	1704	ND		+			
wells 15*	Formation water	Shiiya	1733	96	+	+	+		

* In oil deposits, oil and formation water was collected from several production wells and mix on the ground

Table S2. Analytical method for each molecule dissolved in formation water sample.

Analytical methods	Molecules
Ion chromatography	Cl^-, SO_4^{2-}, Br^-, formate, acetate, propionate, butyrate
Spectrophotometric method using naphthylethylenediamine (NEDA)	NO_2^-
Spectrophotometric method using copper-cadmium reduction NEDA	NO_3^-
Spectrophotometric method using L-ascorbic acid	PO_4^{3-}
Atomic absorption spectrometry	Na^+, K^+, Ca^{2+}, Mg^{2+}
Spectrophotometric method using indophenol blue	NH_4^+
Inductively coupled plasma (ICP) emission spectrometry	Fe, Mn
Combustive oxidation with non-dispersive infrared (NDIR) detection	HCO_3^-, TOC
Spectrophotometric method using zinc acetate precipitation	HS^-
Gas chromatography mass spectrometry	n-alkanes
Spectrophotometric method using 4-aminoantipyrine	Phenols
Gas chromatography mass spectrometry with headspace sampler	Methanol, toluene, xylene

Table S3. Physicochemical characteristics of the formation water samples.

Reservoir	Well no.	Depth	Temp.	pH	ORP	HCO_3^-	Cl^-	SO_4^{2-}	HS^-	NO2⁻	NO_3^-	PO_4^{3-}	Na^+	K^+	NH_4^+	Ca^{2+}	Mg^{2+}	Ba^{2+}	Fe	Mn	TOC	Formate	Acetate	Propio-nate	Butyrate	Methanol	Toluene	Phenols	Xylenes	$\delta^{18}O$	δD
Reservoir	Well no.	mbgs	°C		mV	mg/L	mg/L	mg/L	uM	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mM	mM	mM	mM	μM	μM	mg/L	μM	‰	‰
	well 1	554	38	7.4	-247	720	16000	<5	<0.03	<0.005	<0.005	0.02	9100	180	110	570	190	1.5	4	0.3	18	0.001	0.014	0.01	0.01	1.82	0.22	< 0.2	0.17	-3.83	-14.4
	well 2	653	49	7.3	-230	470	19000	21	0.9	0.011	<0.005	0.044	12000	200	140	780	270	2.2	3.6	0.2	26	0.002	0.002	0.01	0.01					-3	-5.71
	well 3	754	51	7.4	-181	420	19000	<5	0.1	<0.005	<0.005	0.045	12000	220	120	850	200	2.3	4.5	0.4	22	0.001	0.001	0.01	0.01	2.15	0.09	0.3	0.11	-3	-6.09
	well 4	768	56	7.5	-241	520	18000	<5	0.8	0.01	<0.005	0.048	12000	180	100	710	120	1.9	3.6	0.2	31	0.002	0.028	0.03	0.01					-2.34	-5.34
	well 5	883	54	7.3	-230	490	19000	22	0.6	0.01	<0.005	0.043	12000	230	140	830	180	2.5	4	0.2	22	0.001	0.002	0.01	0.01					-3.11	-5.95
Gas-associated aquifers	well 6	798	53	7.4	-215	430	21000	5	1.6	0.01	<0.005	0.066	12000	180	120	730	170	2.3	3.4	0.2	26	0.002	0.007	0.01	0.01					-2.56	-4.67
	well 7	849	58	7.6	-267	500	21000	<5	0.3	<0.005	<0.005	0.1	13000	170	100	700	110	1.9	3.8	0.3	21	0.001	0.002	0.01	0.01	1.76	0.19	0.3	0.14	-2.87	-5.06
	well 8	845	60	7.6	-233	460	21000	<5	0.8	0.011	<0.005	0.104	13000	140	110	610	110	2.4	2.8	0.2	22	0.001	0.002	0.01	0.01					-3.15	-6.09
	well 9	1049	65	7.8	-254	600	20000	6	0.4	0.006	<0.005	0.27	12000	200	120	530	94	4.5	3	0.2	25	0.001	0.013	0.01	0.01	4.76	0.09	0.7	0.14	-2.31	-4.82
	well 10	1115	73	7.5	-290	580	20000	9	2.1	0.009	<0.005	0.35	13000	140	100	320	110	9.5	1.5	0.1	68	0.040	0.900	0.21	0.01	2.62	0.09	3.2	0.13	-2.38	-5.54
	well 11	1373	81	7.7	-205	640	16000	40	2.4	0.007	<0.005	0.32	11000	130	87	240	76	8.6	1.2	<0.1	420	0.430	8.780	1.99	0.17	8.09	0.06	10.8	0.14	-0.64	-6.5
	well 12	1374	82	7.9	-241	480	19000	42	3.6	0.021	<0.005	0.331	12000	140	140	320	99	10	1.1	<0.1	340	0.374	6.152	1.20	0.11					-1.47	-7.05
Oil	wells 13	1513	67	nd	nd	750	10000	31	0.9	<0.005	<0.005	0.007	7500	82	64	140	44	8.5	17	0.2	1500	0.1897	3.725	0.22	nd					-0.55	-12.16
	wells 15	1733	96	6.7	nd	560	16000	31	6.4	<0.005	<0.005	0.01	8900	76	71	200	48	12	75	0.5	1400	1.08	31.77	5.54	1.32	2.99	5.44	9	10.72	0.34	-11.01
# of replication for measurement						1*	1*	1*	3†	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	1*	6‡	6‡

*In the triplicate measurements of the standard sample, the standard deviation was less than 10% of the mean value.

†Standard deviations were less than 20% of each mean value.

‡Standard deviations of δD and $\delta^{18}O$ were less than 2.0 ‰, and 0.5 ‰, respectively.

Table S4. Methanogen diversity based on the mcrA gene in the original formation water and culture samples.

				Original formation water samples						Culture samples						
										Meth/TMA		H_2/CO_2				
			Depth (mbgs)	559	754	845	1049	1115	1373	559	754	559	754	845	1049	1373
			Temp. (°C)	38.2	50.7	57.7	65.2	73	81.1	37	50	37	50	55	65	80
Representative clone ID in OTU	Accession no.	Related species	AA seq. identity (%)	Proportion (%)						Proportion (%)						
N245FW2_mf24	LC783983	*Methanolobus vulcani*	97.9	19.6	59.4	0	2.6	0	0	100	100	0	0	0	0	0
N103FW1_mf25	LC783984	*Methanobacterium alkalithermotolerans*	98.5	47.8	3.1	25	52.6	0	0	0	0	0	18.8	0	0	0
N245FW2_mf32	LC783985	*Methanothermobacter tenebrarum*	96.3	0	15.6	0	0	100	58.6	0	0	0	0	0	0	0
N245FW2_mf25	LC783986	*Methanobacterium ferruginis*	96.2	10.9	21.9	20	2.6	0	0	0	0	13.3	81.3	62.5	0	0
N142FW6_mf24	LC783987	*Methanothermobacter defluvii*	100	0	0	0	0	0	41.4	0	0	0	0	18.8	75	53.3
N103FW1_mf36	LC783988	*Methanobacterium alcaliphilum*	92.6	13	0	45	13.2	0	0	0	0	0	0	0	0	0
N103FW1_mf27	LC783989	*Methanocalculus alkaliphilus*	96.5	4.3	0	5	28.9	0	0	0	0	86.7	0	0	0	0
N142F_HF_m12	LC783990	*Methanothermobacter thermautotrophicus*	99.3	0	0	0	0	0	0	0	0	0	0	18.8	25	46.7
N103FW1_mf37	LC783991	*Methanobacterium alkalithermotolerans*	94.7	4.3	0	5	0	0	0	0	0	0	0	0	0	0

Abbreviation: Meth, methanol; TMA, trimethylamine; AA, amino acids.

Supplementary Figures

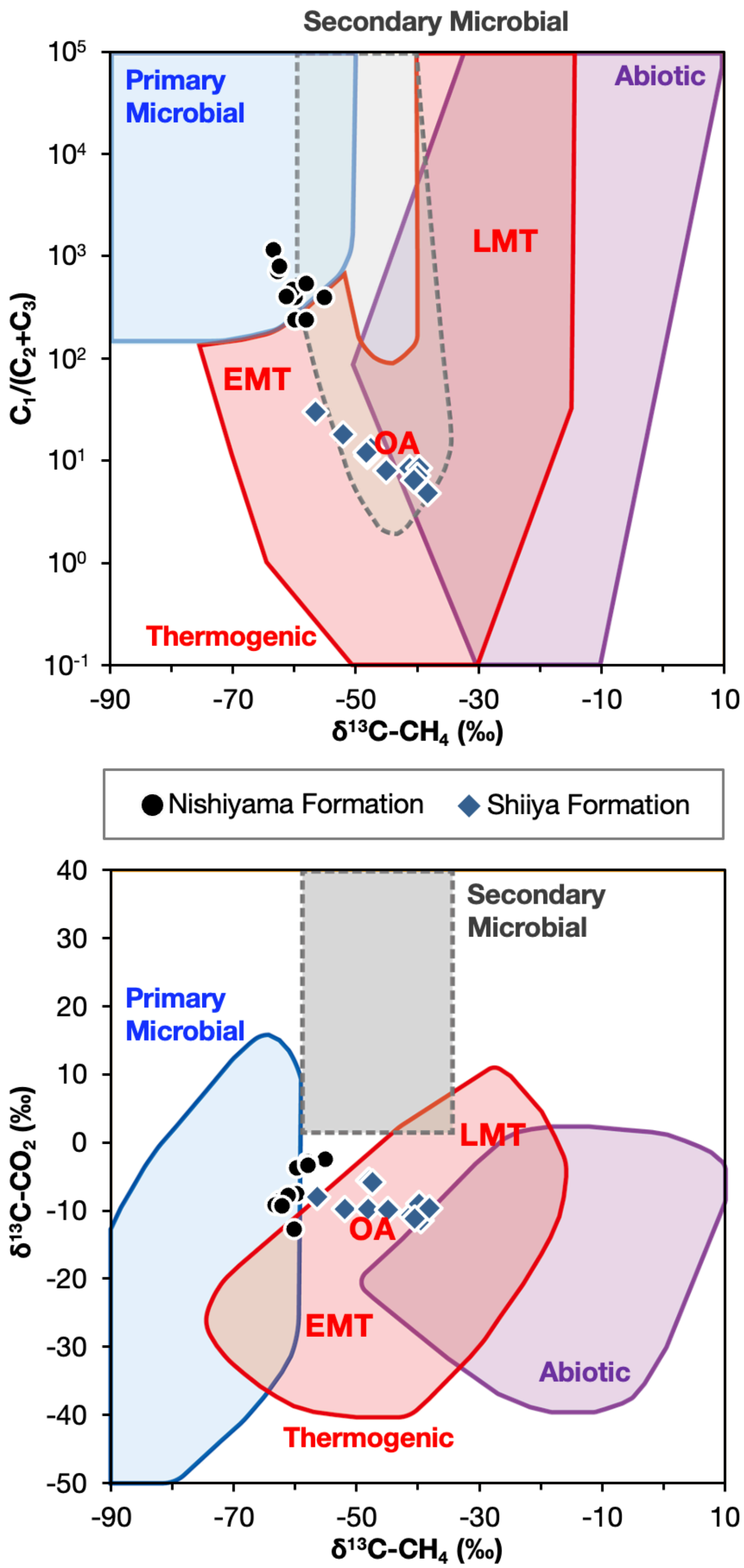

Fig. S1. Genetic diagram of $\delta^{13}C$-CH_4 versus $CH_4/(C_2H_6 + C_3H_8)$ (upper) and $\delta^{13}C$-CH_4 versus $\delta^{13}C$-CO_2 (lower) of gas samples from the Nishiyama and Shiiya Formations. The deduced origin of gas is based on Milkov and Etiope (2018). Abbreviations: EMT, early mature thermogenic gas; OA, oil-associated thermogenic gas; LMT, late mature thermogenic gas.

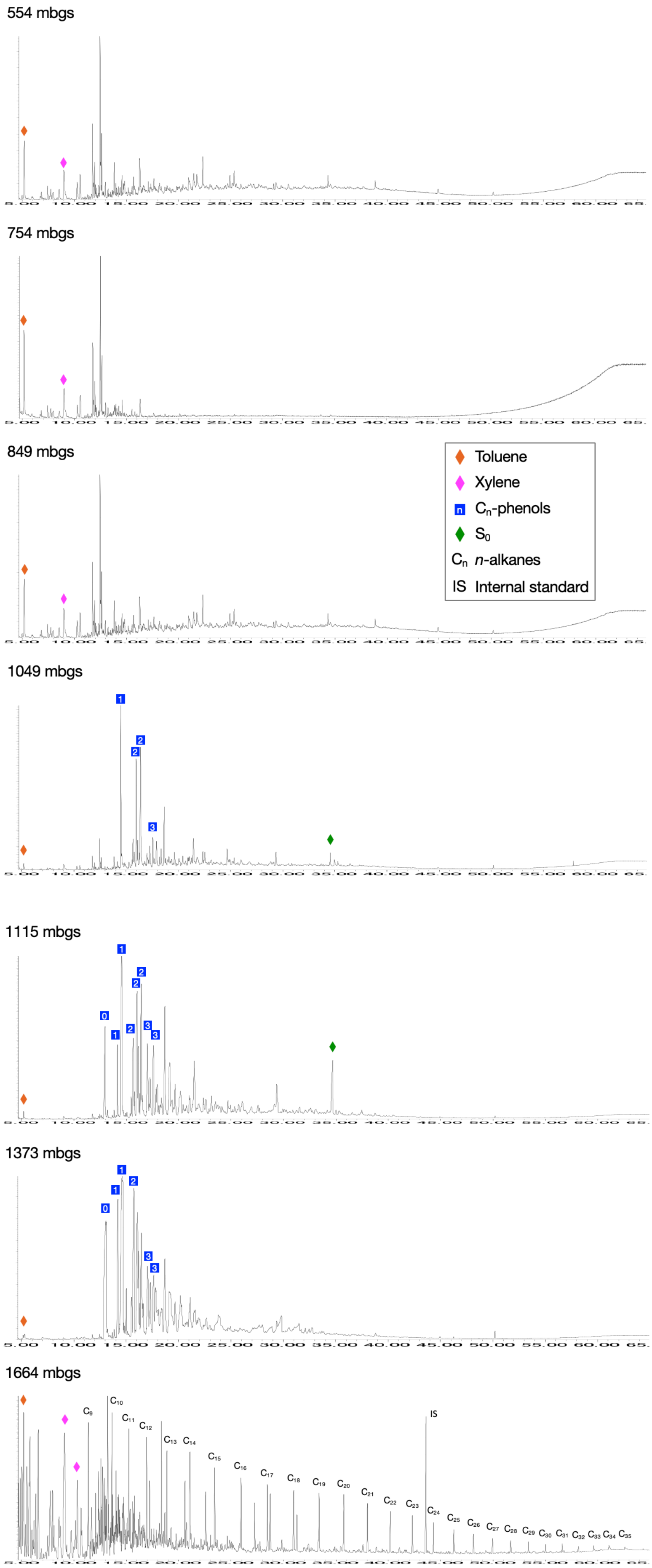

Fig. S2. GC-MS chromatograms of hydrocarbons and phenols in formation water samples and oil sample (1664 mbgs).

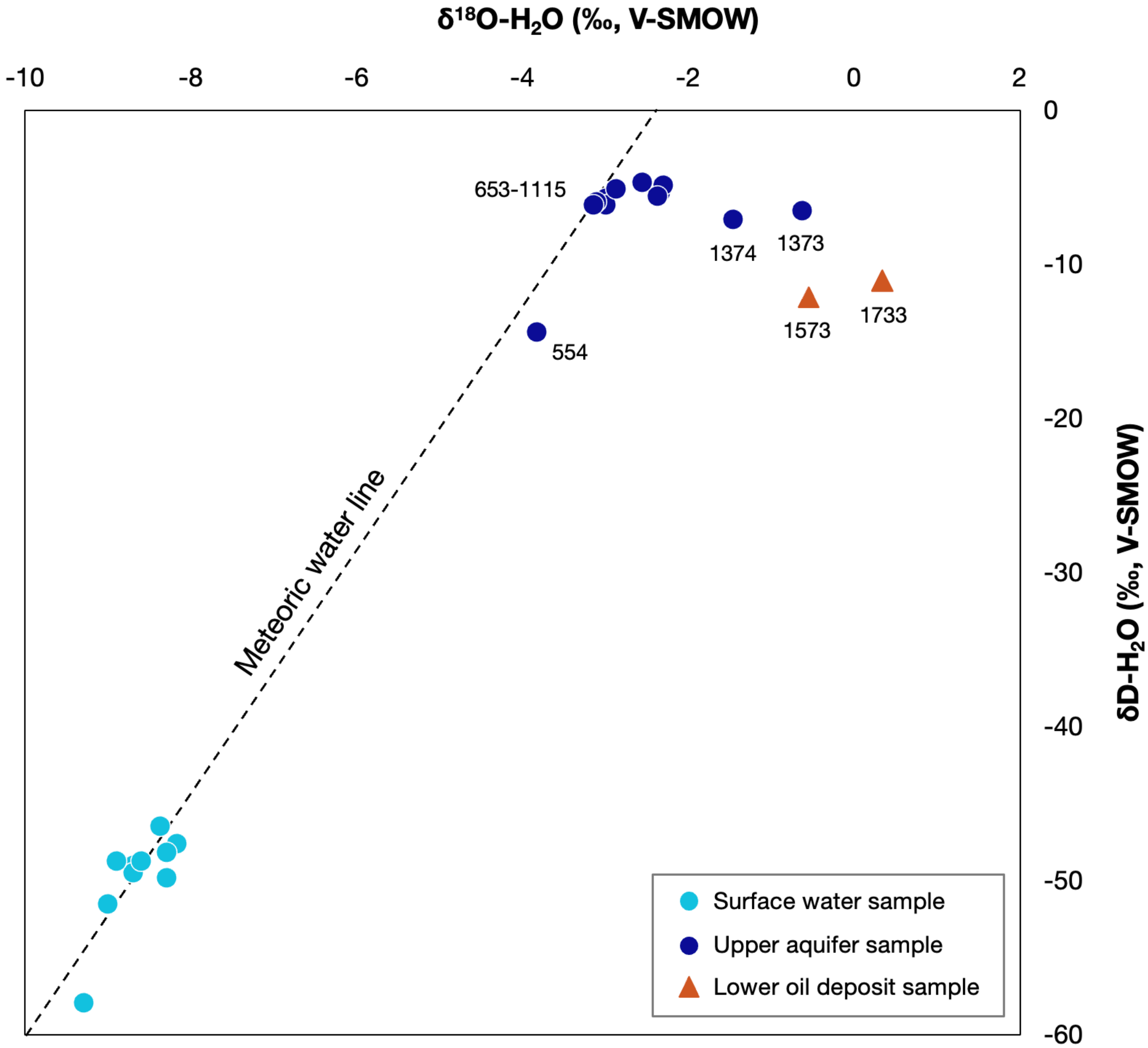

Fig. S3. The δD vs $\delta^{18}O$ plot of formation water samples from upper aquifers and lower oil deposits (numbers indicate average depths), and surface meteoric water in the study area (Waseda and Nakai 1983). The dashed line indicates the meteoric water line in the study area determined by Kato and Kajiwara (1986).

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