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Supplemental Material

Figure S1. Partial gas chromatograms (FID) of hydrocarbon fractions of samples D2-1(A) and T15-1(B) from the Okinawa Trough.

Figure S2. Partial gas chromatograms (FID) of alcohol fractions from samples D2-1(A) and T15-1(B) from the Okinawa Trough.

Figure S3. Partial gas chromatograms (FID) of carboxylic acid fractions of samples D2-1(A) and T15-1(B) from the Okinawa Trough.

Table S1. Contents and stable carbon isotopic composition of hydrocarbons from the Okinawa Trough seep carbonates.

Table S2. Contents and stable carbon isotopic composition of alcohols from the Okinawa Trough seep carbonates.

Table S3. Contents and stable carbon isotopic composition of fatty acids from the Okinawa Trough seep carbonates.

Table S4. The contents of selected major elements, trace elements and enrichment factors of the Okinawa Trough seep carbonates in this study.

Supplementary material for

Formation of authigenic carbonates contributed by sulfate- and metal-driven anaerobic oxidation of methane in the northern Okinawa Trough, East China Sea

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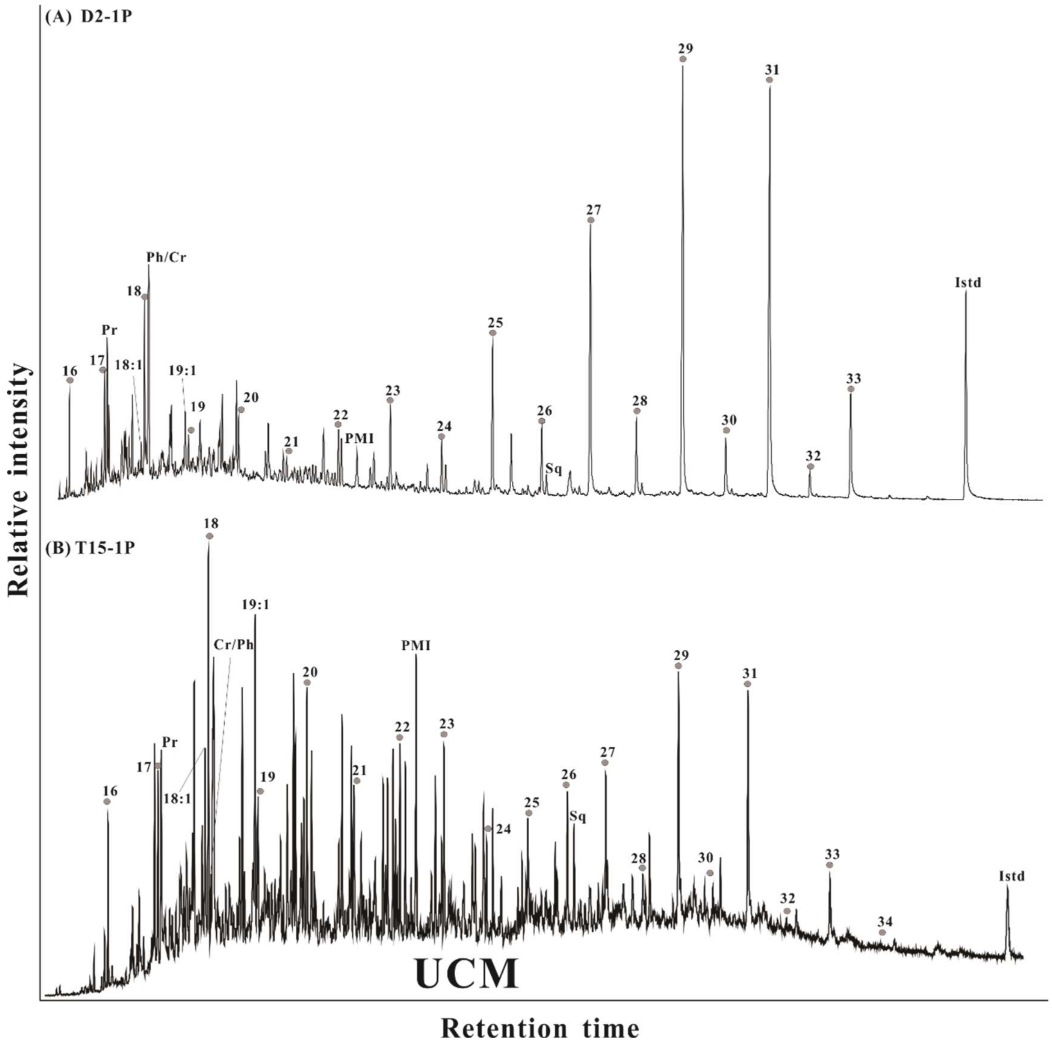

Fig. S1. Partial gas chromatograms (FID) of hydrocarbon fractions of samples D2-1(A) and T15-1(B) from the Okinawa Trough. Istd: internal standard; Pr: pristane; Cr/Ph: crocetane/phytane; PMI: 2,6,10,15,19-pentamethylicosane; Sq: squalane; UCM: unresolved complex mixture; gray dots: *n*-alkanes.

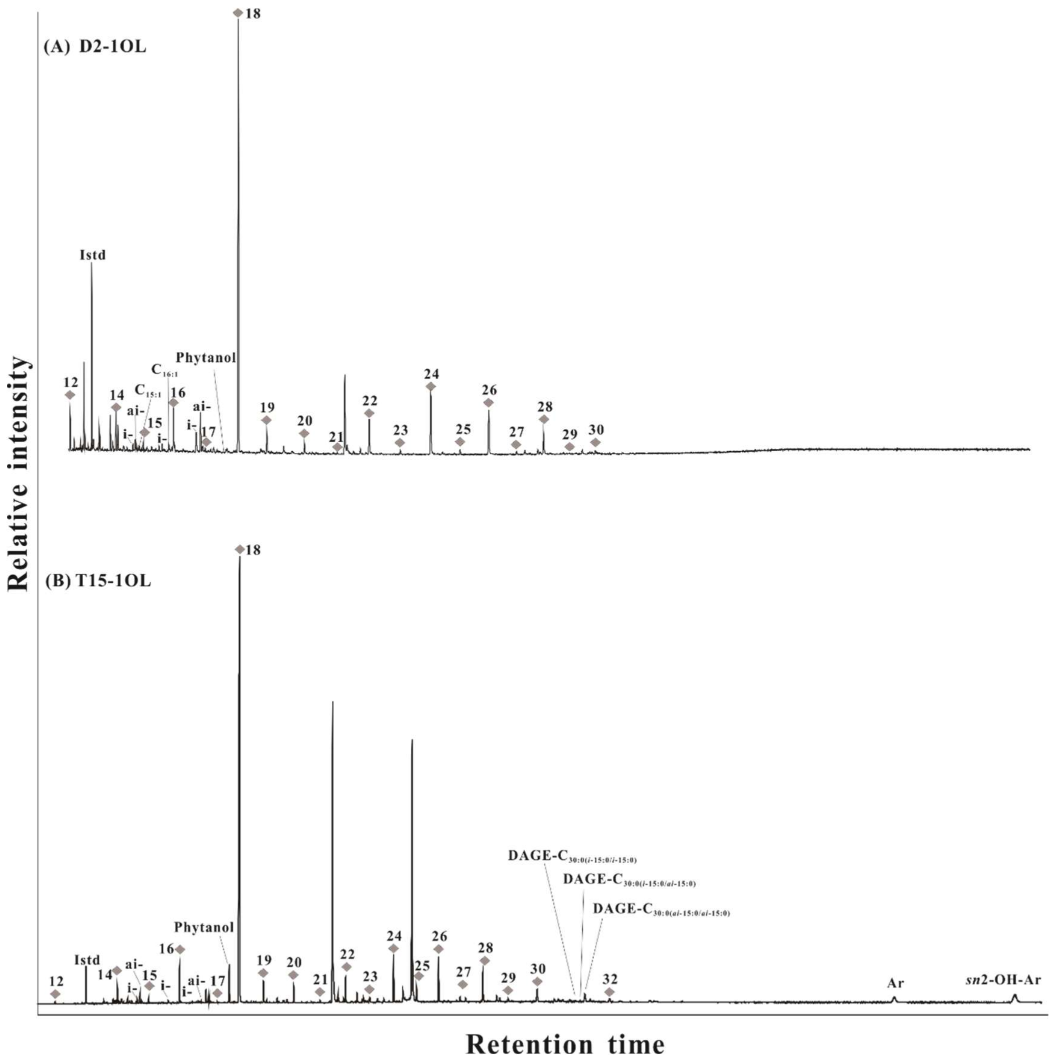

Fig. S2. Partial gas chromatograms (FID) of alcohol fractions from samples D2-1(A) and T15-1(B) from the Okinawa Trough. Istd: internal standard; DAGE: dialkyl glycerol diether; Ar: archaeol; *sn2*-OH−Ar: *sn2*-hydroxyarchaeol; gray diamonds: *n*-alcohols.

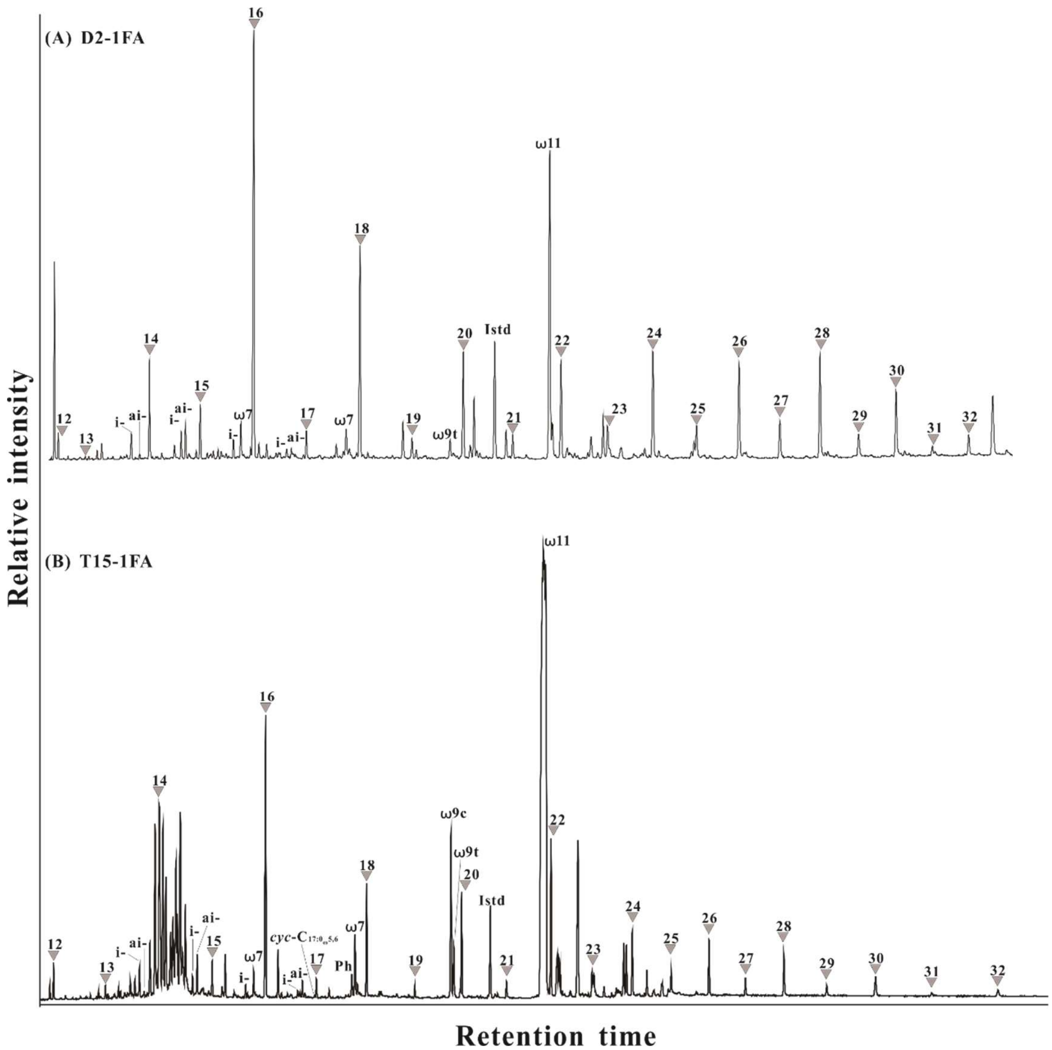

Fig. S3. Partial gas chromatograms (FID) of carboxylic acid fractions of samples D2-1(A) and T15-1(B) from the Okinawa Trough. Istd: internal standard; Ph: phytanoic acid; gray triangles: *n*-fatty acids. ω(c/t): the position of the double bond from the methyl end of the fatty acid, the suffixes c for *cis* and t for *trans* refer to geometric isomers.

Table S1. Contents and stable carbon isotopic composition of hydrocarbons from the Okinawa Trough seep carbonates.

	D2-1		GGD4-1		GGD6		T15-1		T19-3	
	content (ng/g rock)	$\delta^{13}C$ (‰)	content (ng/g rock)	$\delta^{13}C$ (‰)	content (ng/g rock)	$\delta^{13}C$ (‰)	content (ng/g rock)	$\delta^{13}C$ (‰)	content (ng/g rock)	$\delta^{13}C$ (‰)
n–C_{16}	38.1	–30	n.d.	n.d.	17.9	–29	56.7	–29	n.d.	n.d.
n–C_{17}	49.1	–28	0.4	n.d.	15.3	–29	77.5	n.d.	3.5	n.d.
Pr	78.7	–30	0.4	n.d.	9.5	–30	109.7	–31	3.9	n.d.
n–$C_{18:1}$	18.0	n.d.	n.d.	n.d.	n.d.	n.d.	82.1	n.d.	3.5	n.d.
n–C_{18}	101.6	–30	1.3	n.d.	14.7	–29	160.5	n.d.	21.6	–30
Ph[a]	147.2	–36	n.d.	n.d.	13.7	–31	21.9	–44	1.5	–33
n–$C_{19:1}$	42.0	n.d.	n.d.	n.d.	n.d.	n.d.	140.2	n.d.	21.5	n.d.
n–C_{19}	20.1	–31	4.5	n.d.	6.8	–31	67.9	n.d.	20.8	n.d.
n–C_{20}	37.1	–33	14.0	–35	9.4	–29	104.0	n.d.	39.0	–31
n–C_{21}	13.8	–32	17.4	–33	9.0	–31	84.5	–31	37.3	n.d.
n–C_{22}	37.0	–33	16.4	–33	11.8	–32	75.6	n.d.	40.1	–30
PMI	27.1	–111	25.4	–110	1.5	–83	122.0	–118	29.0	n.d.
n–C_{23}	54.8	–29	14.6	–38	14.4	–33	84.5	n.d.	41.1	–38
n–C_{24}	35.9	–32	9.2	–30	9.6	–32	41.7	n.d.	27.3	–33
n–C_{25}	104.1	–31	11.0	–30	7.7	–33	60.8	–30	27.9	–30
n–C_{26}	50.4	–33	9.1	–26	3.9	n.d.	76.6	n.d.	30.7	–30
Sq	13.7	n.d.	3.1	n.d.	1.1	–44	50.1	n.d.	18.1	n.d.
n–C_{27}	210.8	–32	7.8	–32	7.0	n.d.	54.1	–31	26.1	–30
n–C_{28}	56.9	–32	4.4	–29	2.6	n.d.	30.7	n.d.	16.3	–32
n–C_{29}	362.6	–33	10.9	–33	9.6	-34	126.3	–30	51.4	–30

n–C_{30}	45.5	–33	3.4	–29	1.8	n.d.	21.8	n.d.	8.9	–30
n–C_{31}	357.6	–33	9.4	–34	9.7	n.d.	127.6	–32	61.1	–31
n–C_{32}	20.4	–35	2.6	–30	n.d.	n.d.	100.2	–32	4.7	–27
n–C_{33}	86.2	–32	3.1	n.d.	3.3	n.d.	39.1	–32	17.5	–29
n–C_{34}	3.0	–33	n.d.	n.d.	n.d.	n.d.	4.4	n.d.	n.d.	n.d.
n–C_{35}	3.8	–30	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

[a] Co-eluted with crocetane in samples T19-3, T15-1, and D2-1. Pr: pristane; Ph: phytane; Cr: crocetane; PMI: 2,6,10,15,19–pentamethylicosane; Sq: squalane.

Table S2. Contents and stable carbon isotopic composition of alcohols from the Okinawa Trough seep carbonates.

	D2-1		GGD4-1		GGD6		T15-1		T19-3	
	Content (ng/g rock)	$\delta^{13}C$ (‰)	Content (ng/g rock)	$\delta^{13}C$ (‰)	Content (ng/g rock)	$\delta^{13}C$ (‰)	Content (ng/g rock)	$\delta^{13}C$ (‰)	Content (ng/g rock)	$\delta^{13}C$ (‰)
n–C_{14} OL	46.3	–39	13.8	n.d.	9.7	n.d.	79.3	–49	11.0	–39
iso–C_{15} OL	8.3	n.d.	15.1	n.d.	5.3	n.d.	19.4	n.d.	3.3	n.d.
anteiso–C_{15} OL	20.7	n.d.	20.0	n.d.	4.5	n.d.	41.6	–82	4.4	n.d.
n–$C_{15:1}$ OL	8.1	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	6.7	–44
n–C_{15} OL	13.7	n.d.	8.0	n.d.	5.6	n.d.	22.9	n.d.	4.4	n.d.
iso–C16 OL	12.1	n.d.	11.0	n.d.	n.d.	n.d.	10.8	n.d.	n.d.	n.d.
n–$C_{16:1}$ OL	14.5	n.d.	2.7	–43	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
n–C_{16} OL	71.6	–41	66.6	n.d.	40.4	–35	155.5	–54	22.9	–34
iso–C_{17} OL	45.0	n.d.	3.8	n.d.	n.d.	n.d.	5.4	n.d.	2.6	n.d.
anteiso–C_{17} OL	64.2	n.d.	9.2	n.d.	n.d.	n.d.	10.7	n.d.	1.8	n.d.
n–C_{17} OL	10.6	n.d.	8.0	–31	4.2	n.d.	19.9	n.d.	6.1	–30
Phytanol	7.8	n.d.	83.4	–131	5.9	n.d.	151.4	–118	7.1	n.d.
n–C_{18} OL	734.2	–30	536.1	–31	325.5	–30	3870.9	–29	998.3	–29
n–C_{19} OL	41.5	–30	524.5	n.d.	0.0	n.d.	95.5	n.d.	72.3	–30
n–C_{20} OL	22.1	–35	31.5	n.d.	10.3	n.d.	72.7	–38	24.2	–30
n–C_{21} OL	4.7	n.d.	8.7	n.d.	4.7	n.d.	12.5	n.d.	5.8	n.d.
n–C_{22} OL	70.5	–30	79.2	–26	71.0	n.d.	115.1	–23	57.7	–25
n–C_{23} OL	10.7	–26	12.2	n.d.	8.8	n.d.	20.3	n.d.	13.3	–28
n–C_{24} OL	117.5	–31	58.1	–28	50.4	n.d.	207.2	–31	62.1	–28

n–C_{25} OL	10.3	–32	15.2	n.d.	11.9	n.d.	74.1	n.d.	28.0	–32
n–C_{26} OL	92.1	–32	63.1	–30	62.2	n.d.	207.3	–33	71.4	–31
n–C_{27} OL	7.3	–31	8.3	n.d.	10.4	n.d.	26.3	n.d.	12.4	–36
n–C_{28} OL	45.8	–33	110.8	–29	70.3	–32	159.2	–33	77.3	–30
n–C_{29} OL	2.6	–32	100.0	n.d.	10.8	n.d.	23.1	n.d.	16.0	–33
n–C_{30} OL	8.1	–32	50.5	–26	38.4	n.d.	78.2	–28	37.0	–27
DAGE–$C_{30:0(i\text{–}C15:0/i\text{–}C15:0)}$	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	14.4	n.d.	4.7	n.d.
DAGE–$C_{30:0(i\text{–}C15:0/ai\text{–}C15:0)}$	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	10.5	n.d.	3.4	n.d.
DAGE–$C_{30:0(ai\text{–}C15:0/ai\text{–}C15:0)}$	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	61.2	–113	13.2	–104
n–C_{32} OL	n.d.	n.d.	15.2	n.d.	18.2	n.d.	30.5	n.d.	17.0	–27
Ar	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	160.0	–111	3.7	–119
*sn*2–OH–Ar	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	195.3	–107	2.0	n.d.

DAGE: dialkyl glycerol diether; Ar: archaeol; *sn*2–OH–Ar: *sn*2–hydroxyarchaeol

Table S3. Contents and stable carbon isotopic composition of fatty acids from the Okinawa Trough seep carbonates.

	D2-1		GGD4-1		GGD6		T15-1		T19-3	
	Content (ng/g rock)	$\delta^{13}C$ (‰)	Content (ng/g rock)	$\delta^{13}C$ (‰)	Content (ng/g rock)	$\delta^{13}C$ (‰)	Content (ng/g rock)	$\delta^{13}C$ (‰)	Content (ng/g rock)	$\delta^{13}C$ (‰)
n-C_{12} FA	34.9	n.d.	9.9	n.d.	n.d.	n.d.	40.5	n.d.	10.2	n.d.
n-C_{13} FA	6.1	n.d.	9.1	–31	n.d.	n.d.	16.8	n.d.	12.1	n.d.
i-C_{14} FA	50.3	n.d.	10.6	–48	n.d.	n.d.	37.9	n.d.	8.7	n.d.
ai-C_{14} FA	2.8	n.d.	3.4	n.d.	n.d.	n.d.	7.6	n.d.	2.3	n.d.
n-C_{14} FA	166.8	–28	63.0	–35	16.1	n.d.	334.4	n.d.	100.2	–29
i-C_{15} FA	52.6	–28	30.1	–42	4.7	n.d.	20.8	n.d.	25.1	–35
ai-C_{15} FA	59.1	–27	26.6	–72	2.2	n.d.	50.3	–41	25.5	–44
n-C_{15} FA	97.4	–24	42.3	–31	7.8	n.d.	44.9	–37	61.1	–27
i-C_{16} FA	34.2	n.d.	16.9	–50	n.d.	n.d.	17.1	–58	12.7	n.d.
n-$C_{16:1}$ FA	74.0	n.d.	n.d.	n.d.	n.d.	n.d.	41.4	n.d.	6.9	n.d.
n-C_{16} FA	994.3	–30	411.1	–30	311.4	–33	419.8	–30	695.2	–28
i-C_{17} FA	18.6	n.d.	14.1	n.d.	2.8	n.d.	8.0	n.d.	13.9	–35
ai-C_{17} FA	17.8	n.d.	12.7	n.d.	1.4	n.d.	19.5	–77	11.7	–57
n-$C_{17:1}$ FA	n.d.	n.d.	3.5	n.d.	n.d.	n.d.	n.d.	n.d.	2.5	n.d.
n-$C_{17:1}$ FA	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	3.1	n.d.	1.4	n.d.
n-C_{17} FA	58.9	–29	23.2	–29	5.8	n.d.	23.8	–35	40.3	–24
Phytanoic acid	n.d.	n.d.	12.0	n.d.	n.d.	n.d.	30.9	–99	9.4	n.d.
n-$C_{18:1}$ FA	77.6	–29	31.2	–41	29.2	–34	88.1	–25	22.7	–32
n-C_{18} FA	476.3	–30	126.9	–31	97.3	–33	143.3	–35	242.1	–28

n-C_{19} FA	47.6	n.d.	7.1	–34	n.d.	n.d.	15.7	n.d.	13.3	–25
n-$C_{20:1}$ FA	0.0	n.d.	5.9	n.d.	4.6	n.d.	234.3	n.d.	25.6	n.d.
n-$C_{20:1}$ FA	44.6	n.d.	3.2	n.d.	n.d.	n.d.	77.3	n.d.	15.0	n.d.
n-C_{20} FA	244.6	–30	16.6	–41	10.2	n.d.	142.9	–43	58.7	–30
n-C_{21} FA	53.2	n.d.	5.9	–28	n.d.	n.d.	26.3	–27	15.5	–27
n-$C_{22:1}$ FA	812.5	n.d.	276.4	n.d.	62.1	n.d.	2339.2	n.d.	1309.2	n.d.
n-C_{22} FA	230.7	–31	19.8	–34	15.7	n.d.	212.9	–37	91.8	–31
n-C_{23} FA	96.0	–30	6.0	–31	n.d.	n.d.	29.6	n.d.	25.2	–30
n-C_{24} FA	259.2	–31	11.8	–30	5.3	n.d.	94.1	–33	67.4	–26
n-C_{25} FA	76.0	–31	3.6	–23	n.d.	n.d.	37.2	–31	19.5	–24
n-C_{26} FA	233.0	–31	6.5	–32	n.d.	n.d.	78.9	–33	47.2	–27
n-C_{27} FA	97.1	n.d.	1.6	–33	n.d.	n.d.	25.1	–33	10.4	–29
n-C_{28} FA	255.7	–32	3.1	–32	n.d.	n.d.	63.6	–32	21.4	–28
n-C_{29} FA	63.0	–33	n.d.	n.d.	n.d.	n.d.	19.6	–33	4.9	–32
n-C_{30} FA	168.9	–33	n.d.	n.d.	n.d.	n.d.	38.7	–33	6.6	–35
n-C_{31} FA	25.6	–34	n.d.	n.d.	n.d.	n.d.	7.9	n.d.	n.d.	n.d.
n-C_{32} FA	55.8	n.d.	n.d.	n.d.	n.d.	n.d.	13.6	–35	n.d.	n.d.

Table S4 The contents of selected major elements, trace elements and enrichment factors of the Okinawa Trough seep carbonates in this study.

Sample	SiO_2 (%)	Al_2O_3 (%)	CaO (%)	TFe_2O_3 (%)	MgO (%)	P_2O_5 (%)	MnO (%)	Ni (μg/g)	As (μg/g)	Pb (μg/g)	U (μg/g)	Mo (μg/g)	Sb (μg/g)	Ni_{EF}	As_{EF}	Pb_{EF}	U_{EF}	Mo_{EF}	Sb_{EF}	Fe/Al	Mn/Al
D2-1	42.07	5.91	20.2	1.57	4.69	0.14	0.04	23.9	20.4	7.4	4.0	0.39	0.26	0.5	22.8	1.3	1.5	0.7	3.3	0.34	0.01
GGD4-1	35.71	4.58	29.2	1.57	0.74	0.07	0.09	9.3	13.3	8.9	1.6	1.05	0.52	1.8	45.1	1.4	4.7	2.3	8.6	0.44	0.03
GGD6	39.66	6.57	15.40	8.60	5.51	1.13	0.23	47.3	57.7	21.6	3.4	2.58	3.49	2.5	88.9	2.9	2.8	4.0	40.3	1.67	0.05
T15-1	13.06	3.89	38.4	1.68	4.05	0.25	0.15	19.8	17.9	7.3	26.2	2.27	0.48	1.8	46.6	1.7	36.5	5.9	9.4	0.55	0.06
T19-3	31.49	4.91	27.2	2.72	3.78	0.10	0.15	35.1	16.2	9.7	4.3	1.31	0.42	2.5	33.4	1.8	4.7	2.7	6.5	0.71	0.04
D12	27.03	3.98	41.1	1.66	0.85	0.04	0.15	41.7	57.2	59.8	61.7	2.10	4.01	2.1	85.4	7.9	49.4	3.1	44.9	2.66	0.02
GGD16	13.52	6.87	20.1	2.88	4.18	0.18	0.09	47.2	23.2	8.5	8.9	2.17	0.55	4.1	59.0	1.9	12.1	5.5	10.5	0.53	0.06
GGD16-2	40.76	6.78	21.9	14.13	2.06	12.90	0.11	23.4	17.6	11.7	4.1	0.63	0.78	1.2	25.9	1.5	3.2	0.9	8.6	0.53	0.02
GGD4-1-1	41.54	6.27	14.60	6.18	7.25	1.41	0.17	28.6	43.2	16.3	4.9	1.46	1.99	1.6	69.8	2.3	4.2	2.4	24.1	1.26	0.04
GGD4-2	40.83	6.08	16.40	5.48	6.57	0.77	0.08	23.1	32.3	14.1	3.5	0.89	1.83	1.3	53.8	2.1	3.1	1.5	22.9	1.15	0.02
GGD5	50.69	6.02	19.60	1.89	0.86	0.10	0.12	27.4	17.6	12.4	11.4	1.41	0.60	1.6	29.6	1.8	10.3	2.4	7.6	0.40	0.03
ROV07-R2	39.53	6.18	16.65	2.64	8.71	0.35	0.04	18.4	14.6	10.5	9.5	0.74	0.48	1.0	23.9	1.5	8.3	1.2	5.9	0.54	0.01
ROV16	20.92	4.61	27.3	1.85	9.65	0.11	0.12	26.0	17.4	7.3	7.3	1.87	0.44	1.9	38.2	1.4	8.6	4.1	7.2	0.51	0.04
T02	58.04	7.92	9.19	3.04	4.04	0.12	1.34	96.3	16.3	14.1	1.8	12.40	2.18	4.2	20.8	1.6	1.2	15.9	20.9	0.49	0.25
T02-1	60.34	8.11	7.94	3.61	3.95	0.21	0.81	52.3	14.3	15.8	5.5	4.89	1.71	2.2	17.9	1.7	3.7	6.1	16.0	0.57	0.15
T03-1	44.19	7.15	15.25	6.47	4.60	0.22	0.09	46.3	25.8	16.9	2.2	0.74	1.96	2.2	36.5	2.1	1.7	1.0	20.8	1.15	0.02
T03-2	42.78	6.79	14.95	7.36	5.50	0.67	0.10	29.5	33.5	13.7	2.1	0.96	2.14	1.5	50.0	1.8	1.7	1.4	23.9	1.38	0.02
T15	18.24	5.22	34.4	3.03	4.07	0.19	0.04	15.0	18.3	9.8	15.7	0.97	0.71	1.0	35.5	1.7	16.3	1.9	10.3	0.74	0.01
T38	42.49	5.94	15.05	13.03	2.60	4.16	0.13	36.4	69.3	31.2	9.5	1.91	4.55	2.1	118.1	4.7	8.7	3.3	58.2	2.80	0.03

The enrichment factor (EF) were calculated relative to the Earth's upper crust compositions (McLennan, 2001).