Datashare 215

Quantifying sources of gas hydrate through diagenetic modeling

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GEOLOGIC SETTING

The Ulleung Basin

The Ulleung Basin is a back-arc basin located in the southwestern East Sea in a complex tectonic setting that includes the Eurasian, Amur, Philippine Sea, Pacific, and Okhotsk plates (Park et al., 2021). The Ulleung Basin was formed by crustal extension associated with the southward drift of the Japanese Islands Arc away from the Asian mainland during the late Oligocene to Early Miocene (Tamaki et al., 1992; Jolivet et al., 1995). The presence of gas hydrate in the Ulleung Basin has been inferred since the late 1990s (Gardner et al., 1998), and an approximately 0.024 Gt C reserve was estimated to be sealed in gas hydrate (Lee et al., 2013). Authigenic carbonates were reported from the observation of piston cores at 07GHP-10A-1 and ROV07-3&4 (Lim et al., 2009; Nehza et al., 2012).

The Blake Ridge

The Blake Ridge is located at a passive continental margin comprising thinned continental, transitional rift, and oceanic crust mantled by sediments of late Mesozoic and Cenozoic ages (Grow and Markl, 1977; Schlee et al., 1979). Multiple Deep Sea Drilling Project/Ocean Drilling Program (ODP) drilling (i.e., Leg 11, Leg 164, and Leg 172) (Paull and Matsumoto, 2000) have confirmed the occurrence of gas hydrate. The maximum thickness of the gas hydrate stability zone in this region is approximately 700 m along the eastern edge (Lorenson and Collett, 2000), with an estimation of 9 to 52 Gt C (average at 24 Gt C) of methane reserve (Borowski, 2004). Authigenic carbonates were reported from several depth intervals at ODP Leg 164 site 991-997 (Naehr et al., 2000; Pierre et al., 2000; Rodriguez and Paull, 2000).

Gulf of Mexico

The Gulf of Mexico is a passive continental margin along the south coast of North America, opened during the Late Triassic–Early Jurassic (Hudec et al., 2013). Massive gas hydrate has been widely observed and sampled in the northern basins and canyons such as the Orca Basin (Pflaum et al., 1986), Mississippi Canyon (Brooks et al., 1986), and Green Canyon (Phillips et al., 2020; Sassen et al., 2001), with the estimated methane resource held in gas hydrate of 589.3 Gt C (Collett and Kuuskraa, 1998). Abundant authigenic carbonates were found in Atwater Valley (Mansour and Sassen, 2011), Alaminos Canyon (Birgel et al., 2011), and Green Canyon (Roberts et al., 2010).

Bay of Bengal

Prospection of gas hydrate in the Bay of Bengal from the Indian Ocean is implemented mainly in the Krishna-Godavari Basin, the Mahanadi Basin, and the Andaman Basin, with a total gas reserve of 0.19 to 0.48 Gt C (Rastogi et al., 1999). Drilling and coring activities onboard the D/V *JOIDES Resolution* confirmed the presence of gas hydrate associated with vent structures, bathymetric mounds, and vertical and subvertical fractures (Joshi et al., 2019). Active methane gas migration has been reported in the shallow subsurface through the fault system (Ramana et al., 2007, 2009; Dewangan et al., 2010). Few anaerobic oxidation of methane (AOM)-related authigenic carbonates have been reported at Sites 1, 5, 9, 12, and 10 NGHP-01 cores (Teichert et al., 2014; Kocherla et al., 2015).

Table S1. Summary of Molecular and Isotopic Properties of Hydrate-Related Natural Gas

Location	$C_1/(C_2 + C_3)$	$\delta^{13}C_{methane}$, ‰	Average	$\delta D_{methane}$, ‰[a]	Average	Source
Ulleung Basin	463–23,642 (n = 58)	−90.4 to −60.8 (n = 85)	−67.2	−201 to −181 (n = 81)	−193	Choi et al., 2013
	297–6858 (n = 42)	−97.1 to −64.2 (n = 40)	−77.2	−214 to −171 (n = 35)	−200	Kim et al., 2012
	653.4–16,010.1 (n = 25)	−95.4 to −63.6 (n = 19)	−75.6	−219 to −189 (n = 15)	−202	Kim et al., 2011
	109.3–14,706.8 (n = 38)	−88.0 to −60.0 (n = 17)	−76.6	−204 to −190 (n = 10)	−198	Kim et al., 2024
Cascadia Margin	15–46,0317 (n = 386)	−77.1 to −56.8 (n = 412)	−66.4	–		Claypool et al., 2006
	27–11,5882 (n = 58)	−71.5 to −61.2 (n = 60)	−66.4	−208 to −188 (n = 17)	−125	Milkov et al., 2005
	800–205,300 (n = 159)	−84 to −39.5 (n = 160)	−63.4	−186 to −145 (n = 69)	−172	Pohlman et al., 2009
Bay of Bengal	7.3–3945.4 (n = 25)	−85.2 to −61.1 (n = 9)	−72.1	–		Muralidhar et al., 2006
	600–100,898 (n = 194)	−86.6 to −58.9 (n = 222)	−70.2	–		Lorenson and Collett, 2018
	3889–15,926 (n = 55)	−66.5 to −64.5 (n = 55)	−66.2	–		Dixit et al., 2019
	1083.3–3371.9 (n = 4)	−94.7 to −56.7(n = 28)	−72.6	–		Peketi et al., 2022
Blake Ridge	5082–15,088 (n = 6)	−69.7 to −65.9 (n = 6)	−67.0	−206 to −201 (n = 3)	−204	Matsumoto et al., 2000
	970–11,230 (n = 23)	−65.0 to −70.7 (n = 11)	−67.2	−175 to −200 (n = 5)	−190	Lorenson and Collett, 2000
	–	−101.3 to −59.7 (n = 313)	−68.1	−256 to −136 (n = 42)	−187	Paull et al., 2000
	–	−101.3 to −62.4 (n = 25)	−90.4	–		Hoehler et al., 2000
Gulf of Mexico	–	−74.9 to −75.7 (n = 8)	−73.5	−186 to −181 (n = 8)	−184	Burke et al., 1986
	1.9–10,000 (n = 8)	−71.3 to −43.2 (n = 8)	−58.7	–		Brooks et al., 1986
	2400–42,000 (n = 29)	−94.8 to −69.4 (n = 31)	−74.2	–		Pflaum et al., 1986
	1200–420,200 (n = 103)	−84.3 to −70.8 (n = 72)	−73.6	–		Lorenson et al., 2008
	1.7–44.5 (n = 20)	−65.5 to −41.6 (n = 20)	−48.2	−203 to −104 (n = 20)	−158	Sassen et al., 1999
	3–99 (n = 24)	−49.9 to −42.2 (n = 20)	−45.7	−203 to −115 (n = 19)	−175	Sassen et al., 2001
South China Sea	4.1–486.5 (n = 26)	−57.4 to −28.6 (n = 26)	−37.5	–		He et al., 2016
	41.0–35,21.0 (n = 17)	−67 to −47.8 (n = 17)	−50.6	−197 to −170 (n = 17)	−189	Liu et al., 2017
	30.0–69.0 (n = 17)	–		–		Ye et al., 2019
	84.0–2333.0 (n = 8)	−50.1 to −46.5 (n = 8)	−47.5	−178 to −149 (n = 8)	−169	Wei et al., 2018
	5.8–10,000.0 (n = 73)	−74.3 to −46.2 (n = 18)	−60.3	−225 to −180 (n = 4)	−199	Wu et al., 2011
	3.9–70.2 (n = 22)	–		–		Liang et al., 2019
	–	−72.3 to −69.4 (n = 5)	−70.3	−185 to −183 (n = 5)	−184	Zhuang et al., 2016
	7.8–15.1 (n = 12)	−48.6 to −43.2 (n = 4)	−45.2	−221 to −156 (n = 4)	−194	Li et al., 2019
	38.0–42,043.0 (n = 187)	−73.8 to −34.9 (n = 66)	−50.0	−190 to −130 (n = 64)	−137	Zhang et al., 2019
	2.0–3100.0 (n = 20)	−37.8 to −24.0 (n = 25)	−33.5	–	–	Wang et al., 2005
	10.7–9920.0 (n = 114)	−68.2 to −34.4 (n = 114)	−54.7	−198 to −142 (n = 89)	−186	Liang et al., 2022

– = Data not measured or they are unavailable.
[a]The δD values are reported relative to standard mean ocean water.

Table S2. Summary of $\delta^{13}C$ and $\delta^{18}O$ Values of Authigenic Carbonate

Location	$\delta^{13}C$, ‰	Average	$\delta^{18}O$, ‰	Average	Source
Ulleung Basin	−60.7 to −33.9 (n = 7)	−39.3	5.2–5.6 (n = 6)	5.3	Lim et al., 2009
	−43.1 to −27.9 (n = 9)	−37.5	1.1–3.1 (n = 9)	2.7	Nehza et al., 2012
Cascadia Margin	−51.6 to 9.1 (n = 65)	−20.4	−8.7 to 7.7 (n = 65)	4.7	Pierre et al., 2009
	−50.9 to 5.9 (n = 24)	−41.3	−10.8 to 6.5 (n = 24)	3.8	Joseph et al., 2012
	−50.9 to −4.6 (n = 21)	−24.3	−9.3 to 7.3 (n = 21)	3.4	Joseph et al., 2012
Bay of Bengal	−56.0 to 20.0 (n = 123)	−14.8	−1.8 to 6.7 (n = 123)	3.9	Teichert et al., 2014
	−52.4 to −1 (n = 50)	−45.2	3.2–5.2 (n = 50)	4.3	Mazumdar et al., 2009
	−52.4 to 6.4 (n = 88)	−23.7	0.1–6.1 (n = 88)	3.8	Kocherla et al., 2015
	−52.4 to −45.6 (n = 4)	−48.5	4.3–4.8 (n = 4)	4.5	Kocherla et al., 2024
Blake Ridge	−58.5 to 7.8 (n = 152)	−1.9	−5.4 to 7.5 (n = 152)	0.8	Pierre et al., 2000
	−48.4 to −0.4 (n = 111)	−35.2	0.2–5.4 (n = 111)	3.5	Naehr et al., 2000
	−13.79 to 9.4 (n = 153)	0.2	−5.1 to 7.6 (n = 153)	1.6	Rodriguez and Paull, 2000
Gulf of Mexico	−14.6 to 1.1 (n = 114)	−1.3	−5.3 to 1.4 (n = 114)	−2.2	Botz et al., 1988
	−33.9 to −20.4 (n = 32)	−27.9	2.7–5.8 (n = 32)	4.2	Birgel et al., 2011
	−50 to 3.2 (n = 40)	−30.6	2.6–5.9 (n = 40)	4.5	Tong et al., 2019
	−61.9 to −3.1 (n = 35)	−50.3	3.4–5.9 (n = 35)	4.2	Chen et al., 2007
	−28.6 to −9.8 (n = 15)	−23.8	3.7–4.2 (n = 15)	4.0	Mansour and Sassen, 2011
	−29.4 to −1.4 (n = 38)	−13.8	−0.6 to 5 (n = 38)	2.7	Feng et al., 2009
	−31.3 to −0.6 (n = 16)	−21.0	2.8–5.5 (n = 16)	4.0	Feng et al., 2008
	−38.9 to 1 (n = 17)	−12.5	−7.5 to 4.4 (n = 17)	−0.3	Formolo et al., 2004
	−52.3 to −52 (n = 3)	−52.2	4.1–4.5 (n = 3)	4.3	Chen et al., 2005
	−29.8 to −16.1 (n = 37)	−24.1	3.4–5.8 (n = 37)	4.5	Feng et al., 2014
	−36.1 to 5.1 (n = 43)	−14.8	1.9–3.5 (n = 43)	2.8	Bian et al., 2013
South China Sea	−55.3 to −34.3 (n = 47)	−48.5	3.2–4.8 (n = 47)	4.0	Feng and Chen, 2015
	−43 to −27.5 (n = 27)	−39.2	2.5–5.8 (n = 27)	4.8	Liang et al., 2017
	−52.3 to −11.4 (n = 56)	−42.1	1.4–5.1 (n = 56)	3.0	Tong et al., 2013
	−49.2 to 2.4 (n = 7)	−23.1	0.4–3.7 (n = 7)	2.7	Wang et al., 2012
	−47.8 to −20.5 (n = 15)	−40.2	0.5–3.8 (n = 15)	2.3	Lu et al., 2014
	−62.3 to −48.5 (n = 19)	−55.6	3.4–6 (n = 19)	4.4	Deng et al., 2019
	−46.8 to −22.9 (n = 49)	−38.3	−2.8 to 3.5 (n = 49)	2.0	Yang et al., 2018
	−57.6 to −35.7 (n = 60)	−50.0	1.7–5.3 (n = 60)	3.8	Han et al., 2008
	−51.8 to 0.4 (n = 5)	−30.8	0.4–5.1 (n = 5)	3.2	Chen et al., 2005
	−30.6 to −0.3 (n = 7)	−24.3	3.1–3.6 (n = 7)	3.4	Wang et al., 2014
	−15.9 to −1.4 (n = 14)	−6.3	2.7–3.8 (n = 14)	3.4	Wan et al., 2018
	−43.5 to −33.8 (n = 32)	−40.3	0.5–3.8 (n = 32)	3.3	Wei et al., 2023
	−43.5 to −25.4 (n = 96)	−34.6	0.5–4.9 (n = 96)	3.7	Liu et al., 2022

The Cascadia Margin

The Cascadia margin is an accretionary margin, grown by accretion of various terranes during Cenozoic subduction (Bohrmann et al., 1998). Gas hydrate has been found in this region, including ODP Leg 146 and Leg 204 (Tréhu, 2003), with the estimated methane resource held in gas hydrate up to 1.6×10^{-5} Gt C (Milkov et al., 2005). Cold seeps have been recorded in numerous localities (Torres et al., 2010), with a methane flux rate of approximately 3×10^{4} mol h^{-1} (Heeschen et al., 2005). The AOM-related authigenic carbonates were discovered in these hydrates, in International Ocean Discovery Program Sites U1325–U1329 (Blanc-Valleron et al., 2009; Pohlman et al., 2009).

The South China Sea

The South China Sea (SCS) is one of the largest marginal seas in the western Pacific (Zhang et al., 2016),

Table S3. U-Th/^{14}C Age and δ^{13}C Values of Authigenic Carbonate

Location	Type	Age, ka	δ^{13}C, ‰	Source
Ulleung Basin	^{14}C	10.8–24.3 (n = 4)	–	Bahk et al., 2013
Cascadia Margin	U-Th	0.8–267.6 (n = 26)	–	Teichert et al., 2003
Bay of Bengal	^{14}C	40.1–51.6 (n = 5)	−46 to −2.5 (n = 5)	Teichert et al., 2014
	^{14}C	0.5–43.6 (n = 16)	−52.4 to −45.1 (n = 7)	Mazumdar et al., 2009
Blake Ridge	^{14}C	0.5–26.9 (n = 6)	1.1–2 (n = 6)	Rodriguez and Paull, 2000
Gulf of Mexico	^{14}C	6.7–22.5 (n = 6)	−25 to −1.3 (n = 6)	Feng et al., 2008
	^{14}C	1.2–46.5 (n = 15)	−2.9 to 3.8 (n = 15)	Bian et al., 2013
	U-Th	1.6–53.4 (n = 20)	−51.8 to −28.1 (n = 20)	Feng et al., 2010
	U-Th	1.4–194.9 (n = 15)	−53.3 to 2.8 (n = 14)	Aharon et al., 1997
South China Sea	^{14}C	1.9–10.6 (n = 9)	−20 to −2.3 (n = 9)	Feng and Chen, 2015
	^{14}C	2.4–8.2 (n = 5)	0.6–2.8 (n = 5)	Hu et al., 2017
	^{14}C	2.9–6.1 (n = 11)	−4.5 to 0.7 (n = 11)	Liang et al., 2017
	^{14}C	1.9–40.6 (n = 9)	−2.4 to 0.6 (n = 9)	Wan et al., 2018
	U-Th	63–330 (n = 10)	−51.5 to −32.6 (n = 9)	Tong et al., 2013
	U-Th	11.5–144.5 (n = 15)	−56.7 to 2.7 (n = 15)	Han et al., 2014
	U-Th	27.7–55 (n = 7)	−43.4 to −22.9 (n = 7)	Yang et al., 2018
	U-Th	112.7–133.3 (n = 41)	−53 to −40.5 (n = 41)	Chen et al., 2019
	U-Th	12.2–136.3 (n = 3)	−38.1 to −26.9 (n = 3)	Wei et al., 2020

– = Data not measured or they are unavailable.

Table S4. Summary of ^{87}Sr/^{86}Sr Values of Authigenic Carbonate

Location	^{87}Sr/^{86}Sr	Source
Cascadia Margin	0.705420–0.709206 (n = 24)	Joseph et al., 2012
Blake Ridge	0.709125–0.709206 (n = 14)	Naehr et al., 2000
South China Sea	0.709025–0.709259 (n = 20)	Tong et al., 2013

which covers an area of 3.5×10^6 km^2 (Wang et al., 2018). The drilling exploration confirmed that the gas hydrates in the SCS were preferentially found at the northern passive continental margin within relative shallow burial depths, with a total inventory reserve of approximately 42.8 Gt C (Wang et al., 2018). Massive authigenic carbonates were discovered from a relatively shallow burial depth (<22.6 m below seafloor) at an active cold seep region and/or associated with gas hydrate (i.e., the Haima cold seep and the GMGS08 in the Dongsha Area) (Chen et al., 2016).

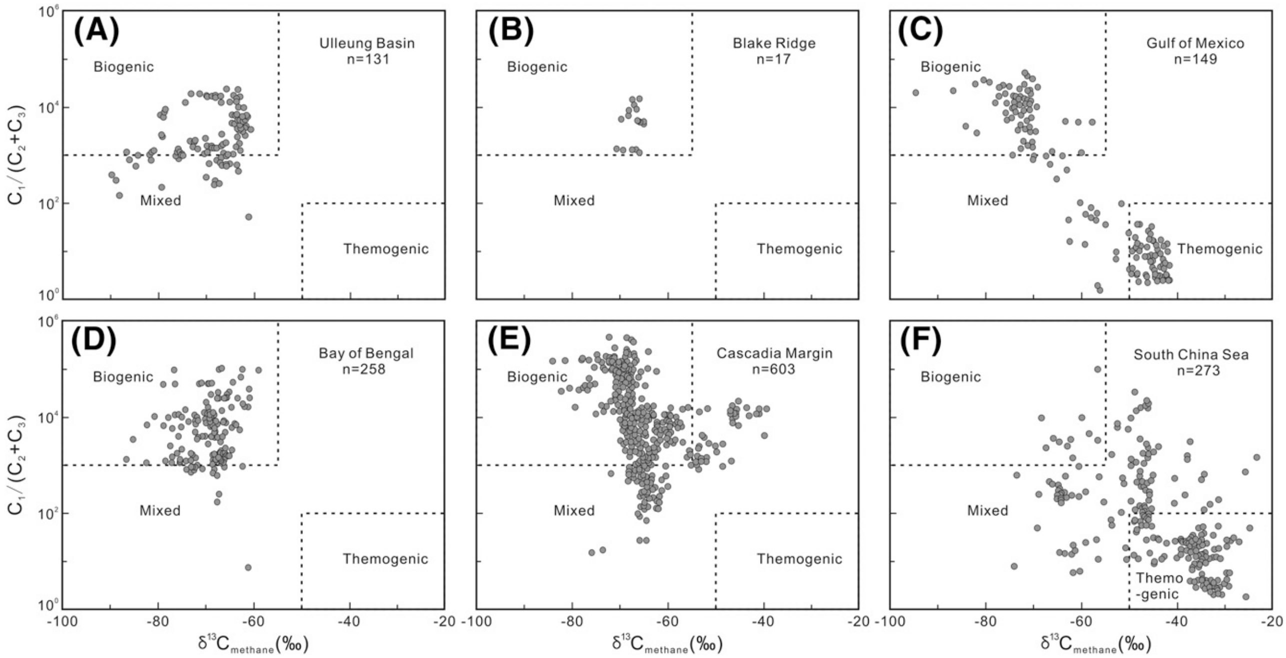

Figure S1. $C_1/(C_2+C_3)$-$\delta^{13}C_{methane}$ relationship of hydrate-associated gas from study areas. (Data and their sourced references were listed in Table S1).

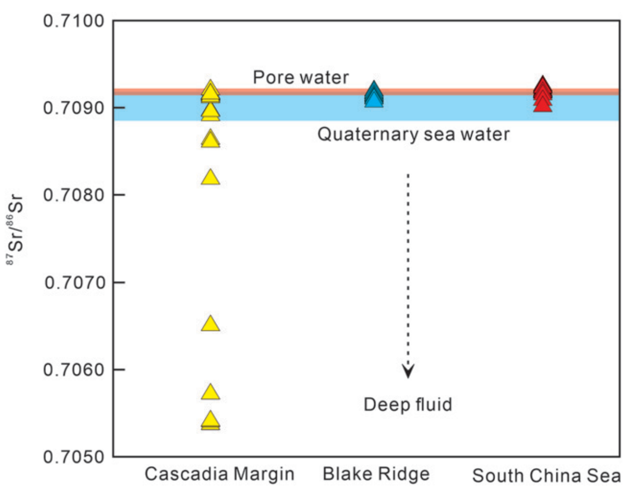

Figure S2. Plot of $^{87}Sr/^{86}Sr$ ratios of authigenic carbonates showing their comparison with those of Quaternary seawater (Veizer et al., 1999) and pore water from the Blake Ridge (Naehr et al., 2000).

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