Table 1. Brief summary of the origins and sources of hydrate bound-gases around the world published in the past.

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| --- | --- | --- | --- | --- |
| **Region** | **Location** | **Reference** | | **Brief description of origins and sources of hydrate-bound gases** |
| South China  Sea | Shenhu | (Fu and Lu, 2010)  (Liu et al., 2015b)  (Dai et al., 2017) | | Methane of hydrate-bound gas is a microbial origin produced by CO2 reduction. |
| (Ye et al., 2018) | | Methane of the natural gas hydrate in the Shenhu area was mainly derived from the bacterial reduction of CO2, but the contribution of the thermogenic origin cannot be excluded. |
| (Zhang et al., 2019) | | Mixed biogenic-thermogenic origin for the hydrate-forming gas. The methane isotope correlation indicates that the source of the hydrate gas is closely related to the deep conventional gas reservoirs discovered in the Baiyun Sag-Panyu Low Uplift area. |
| (Liang et al., 2022) | | Hydrate-bound gases have a mixed origin, containing both biogenic and thermogenic gases. Biogenic and thermogenic hydrocarbons were derived from marine organic matter and terrestrial organic matter, respectively. |
| (Lai et al., 2022) | | Sites SC1 and SC2 exhibit the geochemical characteristics of secondary microbial gas, and the propane (C3) component and liquid thermogenic hydrocarbons (C15+) have been severely biodegraded. Secondary microbial methane within gas hydrates is an important terminal product that has been converted from thermogenic hydrocarbons via microbial degradation |
| Qiongdongnan | (Lai et al., 2021) | | The stable carbon isotopic compositions of C1–C3 hydrocarbon gas components indicate that the methane has a mixed microbial and thermogenic origin, while the C2+ hydrocarbon gases are of thermogenic origin derived from coaly-type source rocks. |
| (Fang et al., 2019) | | Gases are dominated by methane with small amounts of ethane and propane and had relatively light δ13C-CH4, indicating mixtures of biogenic and thermogenic gas. |
| (Ye et al., 2019) | | Thermogenic origin for the gas. |
| (Wei et al., 2021) | | Mixture of microbial and thermogenic gases. |
| Taixinan | (Liang et al., 2017) | | The gas source of hydrate is mainly microbial gas, cracking gas may provide gas source together with microbial gas in the form of biodegradable gas. |
| (Sha et al., 2019) | | Microbial gas. |
| Okinawa Trough |  | (Li et al., 2021) | | Methane having a mixture of thermogenic and microbial sources. |
|  | (Xu et al., 2021) | | Active seepage of biogenic and thermogenic methane was identified on fault scarps and dome structures, respectively. |
| Qilian Mountain permafrost |  | (Lu et al., 2010) | | The gas of natural gas hydrate is mainly thermogenic origin (mainly related to oil cracking) with a small amount of microbial origin (acetate fermentation). |
|  | (Lu et al., 2013) | | Gases from gas hydrate is mainly concomitant with deep oil or crude oil in the study area. Strata within gas hydrate stability zone play little role in gas sources for gas hydrate. |
|  | (Cheng et al., 2018) | | Organic thermogenic gas derived from Middle Jurassic source rocks. |
|  | (Wang et al., 2015b) | | Most of the hydrate-bound gases are thermogenic in origin, with minor additional mixed microbial and thermogenic methane that is sourced from sapropelic kerogens in the underlying hydrocarbon reservoir. |
|  | (Liu et al., 2015a) | | Hydrate-bound gases are from thermogenic origin. |
|  | (Dai et al., 2017) | | Gases of the gas hydrate samples from the Jurassic Jiangcang Formation in the Muli County in Qilian Mountain are mainly of oil-derived origin, characterized by self-generation and self-preservation. |
| Offshore India | KG Basin and Andaman Site | (Stern and Lorenson, 2014) | | The hydrate-forming gas is predominantly methane with trace quantities of higher molecular weight hydrocarbons of primarily microbial origin. |
|  | (Lorenson and Collett, 2018) | | Hydrate from the Krishna-Godavari Basin is mainly microbial methane. Gas from the Mahanadi Basin was mainly methane with microbial gas source; however deeper cores contained higher molecular weight hydrocarbon gases suggesting a small contribution from a thermogenic gas source. Gas composition in the Andaman Basin was mainly microbial gas sources. |
|  | (Dixit et al., 2019) | | Carbon isotopic studies show that methane gas sampled from recovered gas hydrate samples were derived from biogenic (microbial) sources. |
| Krishna-Godavari Basin | (Kida et al., 201i9) | | The gas hydrate is structure I with hydration number n=6.1–6.2, formed predominantly from microbial methane and small amounts of heavier hydrocarbons up to C3. |
| South of Pakistan | Markeran accretionary prism | (Lalk et al., 2022) | | Δ13CH3D values consistent with a microbial source of methane, produced between 46 and 65 °C. |
| Offshore Korea | Ulleung Basin | (Kim et al., 2011) | | Microbial CO2 reduction. Methane source of the hydrate-bound gas is the same as that of headspace and void gas. |
| (Choi et al., 2013) | | Methane predominantly originates via microbial carbon dioxide reduction. |
| (Kim et al., 2013) | | Microbial source for the CH4 and C2H6. |
| Offshore Japan | Nankai Trough | (Kida et al., 2015) | | Microbial origin for the natural gas distributed at this site |
| Nankai accretionary complex | (Ijiri A. et al., 2018) | | Clumped methane isotopologues suggest that ~90% of methane is microbially produced at 16° to 30°C |
| Joetsu Basin | (Hachikubo et al., 2015) | | The hydrate-bound hydrocarbons at Umitaka Spur (southwestern Joetsu Basin) primarily consisted of thermogenic methane, whereas those at Joetsu Knoll contained both thermogenic methane and a mixture of thermogenic and microbial methane. |
| (Snyder et al., 2020) | | The presence of hydrate with a mantle 3He/4He signatures indicates that fluids enriched in mantle gases are responsible for the mobilization of thermogenic gas within active Japan Sea chimney structures. |
| (Zhang et al., 2021) | | By combining clumped isotope results with other traditional approaches, a thermogenic and two microbial end-members as well as their isotopic compositions were identified and the relative contribution of each end-member was also quantified. |
| Kumano Basin | (Lalk et al., 2022) | | High Δ13CH3D values (>4.7‰) are consistent with a shallow microbial source. |
| Sea of Okhotsk | Offshore Sakhalin Island | (Hachikubo et al., 2010b) | | Methane was produced by microbial reduction of CO2. Small amounts of thermogenic gas were mixed with microbial methane. |
| Northwest Atlantic | Blake Ridge | (Ryo Matsumoto, 2000) | | Methane was generated through bacterial CO2 reduction. |
| (Lorenson, 2000) | | Methane of this isotopic composition is mainly microbial in origin and likely produced by bacterial reduction of bicarbonate. The hydrocarbon gases here are likely the products of early microbial diagenesis. |
| Alaska North Slope | Mallik | (Lorenson et al., 1999) | | In situ gases can be divided into three zones composed of mixtures of microbial and thermogenic gases. Thermogenic gas likely migrated from depths below 5000 m. |
| Mount Elbert | (Lorenson et al., 2011) | | These results are consistent with the concept that the Eileen gas hydrates contain a mixture of deep-sourced, microbially biodegraded thermogenic gas, with lesser amounts of thermogenic oil-associated gas, and coal gas. Thermal gases are likely sourced from existing oil and gas accumulations that have migrated up-dip and/or up-fault and formed gas hydrate in response to climate cooling with permafrost formation. |
| Mount Elbert | (Stern et al., 2011) | | These gas composition results are also consistent with gas hydrate sampled by direct dissociation in plastic syringes on the drill site (Lorenson et al., 2011). |
| Cascadia Margin | Hydrate ridge | (Milkov et al., 2005) | | Two end-member gas sources (deep allochthonous and in situ) as mixtures of different proportions. In an area of high gas flux gas hydrates are composed of mainly allochthonous mixed microbial and thermogenic gases, while areas with low gas flux are mainly from microbial methane and ethane generated dominantly in situ. |
| (Winckler et al., 2002) | | The hydrates contain no He and Ne, but contain large amount of Ar, Kr and Xe, indicating that light noble gases are not incorporated into the hydrate structure. Microbial CO2-reduction is the dominant CH4 production pathway. |
| (Claypool, 2006) | | The near-surface gas hydrates are mainly composed of previously buried microbial methane but also contain a significant (10–15%) component of thermogenic gases and are overprinted with microbial methane generated currently in shallow sediments. |
| (Lalk et al., 2022) | | Clumped methane isotopologues suggest that ~90% of methane is microbially produced at 18°C to 34°C |
| (Wang et al., 2015a) | | Microbial methane in pore waters and gas hydrates from northern Cascadia margin sediments yielded Δ13CH3D temperatures of 12° to 42°C. These are consistent with their expected low formation temperatures. |
| Barkley Canyon | (Pohlman et al., 2005) | | Thermogenic gas source. The source rock for the Barkley Canyon hydrate and vent gas had primarily Type III kerogen mixed with a small fraction of Type II kerogen. |
| (Lu et al., 2007) | | The sample contains structure H hydrate with thermogenic origin |
| Offshore Costa Rica | Middle America Trench | (Lückge et al., 2002) | | Most of the gas was generated by microbial CO2-reduction. Percentage of thermally-generated ethane increases with depth. |
| Gulf of Mexico | Mississippi Canyon | (Sassen et al., 2001a) | | The isotopic properties of C1±C5 gas from reservoirs, vents, and gas hydrate correlate closely. Free hydrocarbon gas, gas hydrate, and authigenic minerals in chemosynthetic communities of the northern Gulf of Mexico continental slope: relation to microbial processes |
| Chapopote Knoll, | (Klapp et al., 2010) | | Predominantly fueled by thermogenic hydrocarbon sources. |
| Campeche Knolls | (MacDonald et al., 2004) | | Molecular and isotopic compositions of the gas hydrate and sediment headspace from the second grab sample indicate moderately mature, thermogenic gas. |
| Atwater Canyon | (Sassen et al., 2001b) | | A leaky petroleum system is proposed to be the main source of thermogenic gases. |
| Atwater Canyon | (Sassen et al., 1999) | | Most thermogenic gas hydrate occurrences in the central slope are linked to vertical migration of oil and gas from deep Upper Jurassic source rock facies. Bacterial methane hydrates also occur. |
| Gulf of Mexico | (Lalk et al., 2022) | | Mixing between estimated microbial and thermogenic endmembers show that oil-associated hydrates from the Gulf of Mexico may be 70 to 80% thermogenic in origin. Δ13CH3D temperatures range from 115° to 118°C. |
| Brazil’s Continental Margin | Rio Grande Cone; Amazon deep-sea fan | (Ketzer et al., 2019) | | Dominantly formed by biogenic methane. |
| Amazon Deep-Sea Fan and Slope Sediments | (Rodrigues et al., 2019) | | Dominant microbial origin of methane via carbon dioxide reduction. However, a mixture of thermogenic and microbial gases was suggested for the hydrate-bound and dissolved gases in the continental slope adjacent to the Amazon fan. |
| Amazon deep-sea fan | (Ketzer et al., 2018) | | Gas compositions from hydrates recovered in vents at three locations on and north of the fan indicate biogenic sources, whereas samples from vents adjacent to the fan proper include possible thermogenic contributions. |
| Rio Grande Cone | (Miller et al., 2015) | | The chemical and isotopic compositions of the gas strongly suggest a biogenic origin for the analyzed samples. |
| Chukchi Sea |  | (Kim et al., 2020) | | The isotopic signatures of gas samples at the hydrate-bearing sites and below the SMT at the nonhydrate-bearing sites reflect thermogenic source transported across at least 1 km through faults/fractures in the Chukchi Sea. |
| Norwegian Sea | Nyegga pockmark | (Vaular et al., 2010) | | Methane is microbially produced and originates from CO2 reduction. Ethane from the gas hydrate has mixed thermogenic and microbial contributions, which is supported by the existence of propane and isobutane. |
| (Hovland et al., 2005) | | Presence of both bacterial and thermogenic gases. |
| Barent sea | North Atlantic Haakon Mosby | (Lalk et al., 2022) | | Microbial-like C1/C2+ C3, Δ13CH3D, and δD-CH4. The values of Δ13CH3D from these sites, however, are low (ca. < 3.5‰, T13D range from 264°C to 313°C). The tectonic settings of the mud volcanoes may have important implications for chemistry of their deeply sourced fluids and mechanism of methane production. |
| Black Sea | Dvurechenskii mud volcano | (Blinova et al., 2003) | | Mainly biogenic origin with an admixture of thermogenic gas. |
| Batumi seep | (Pape et al., 2010a) | | Predominant microbial methane formation. |
| Sorokin Trough | (Stadnitskaia et al., 2008) | | Gases are initially derived from the comparable hydrocarbon pools and are likely initial products of non-microbial oil cracking processes. Dry characteristics of gas and 13C-depleted signatures of methane are result of a high admixture of secondary microbial gas formed due to subsequent microbial anaerobic degradation of redeposited hydrocarbons in the shallow reservoirs. |
| Black Sea | (Lalk et al., 2022) | | Gas hydrate from mud volcanoes located on km-thick sediments in tectonically less active or passive settings yielded microbial-like δ13C-CH4 and C1/C2+C3 values, and low Δ13CH3D values (1.6–3.3‰), which may be due to kinetic isotope effects. |
| Sea of Marmara |  | (Bourry et al., 2009) | | The gas bubbles contain thermogenic methane and are likely sourced from the natural gas reservoirs of the Thrace Basin, or from their source rocks. Gas bubbles from Central High also show a thermogenic origin whereas gas bubbles sampled on the Çinarcik Basin are composed of biogenic methane, mixed with a trace amount of thermogenic ethane |
|  | (Ruffine et al., 2018) | | Microbial sources producing methane from primary methanogenesis have been identified in the Tekirdağ and the Çınarcık basins. In addition, six different thermogenic reservoirs or migration pathways are responsible for the supply of gas to the seeps on the highs and in the western basin. Five of them are undergoing biodegradation followed by secondary methanogenesis, thereby providing additional sources of microbial methane to the seeps. |
|  | (Giunta et al., 2021) | | Isotopic characteristics of methane appears to be affected to varying degrees by bond re-equilibration with clay minerals, which might imply that the temperature obtained from clumped isotope represents the re-equilibration condition of post-generation rather than the actual formation temperature of methane. |
| Mediterranean Sea | Amsterdam mud volcano | (Pape et al., 2010b) | | Prevalence of thermogenic light hydrocarbons. |
| Olimpi Mud Volcano field; Anaximander Mountains | (Charlou et al., 2003) | | Methane to ethane ratios (>1000) and δ13C-CH4 values (-65.6‰ PDB) indicate that the CH4 is microbially produced. |
| Mediterranean Sea | (Lalk et al., 2022) | | Typical δ13C-CH4 values for thermogenic sources, while Δ13CH3D values (3.8–6.0‰) consistent with prevailing microbial sources (15–59°C). |
| Offshore South Iberia & NW Africa Margin | Gulf of Cadiz (Ginsburg Mud volcano) | (Mazurenko et al., 2002) | | The inferred source of the gas in the hydrates is enriched in C2–C6 (≤5%), indicating that the gas has a thermogenic origin. |
| West African | Congo–Angola Basin | (Charlou et al., 2004) | | Primarily microbial origin for the CH4, which is generated through bacterial CO2 reduction |
| Northern Congo Fan | (Lalk et al., 2022) | | Δ13CH3D values consistent with a microbial source of methane, produced between 39 and 54 °C. |
| Gulf of Guinea | (Lalk et al., 2022) | | Δ13CH3D values consistent with a microbial source of methane, produced between 36 and 54 °C. |
| Offshore New Zealand | Hikurangi subduction margin | (Koch et al., 2016) | | The analyses clearly show that the cold vents at Opouawe Bank (as well as at the Hikurangi margin in general) are fueled by the seepage of biogenic methane gas. |
| Lake Baikal | Malenky, Bolshoy and K-2 mud volcanoes | | (Kida et al., 2009) | Microbial origin by methyl-type (acetic) fermentation |
| Kukuy Canyon | (Kida et al., 2006) | | Gas composition and crystallographic analyses of hydrate samples reveal involvement of two distinct gas source types in gas hydrate formation at present or in the past: microbial (methane) and thermogenic (methane and ethane) gas types. The clathrate structure II, observed for the first time in fresh water sediments, is believed to be formed by higher mixing of thermogenic gas. |
| Lake Baikal | (Hachikubo et al., 2010a) | | All the seep sites are with dominant microbial origin of methane via methyl-type fermentation; Two sites are with mixture of thermogenic and microbial gases |

Blinova, V.N., Ivanov, M.K., and Bohrmann, G. (2003). Hydrocarbon gases in deposits from mud volcanoes in the Sorokin Trough, north-eastern Black Sea. *Geo-Marine Letters* 23(3-4)**,** 250-257. doi: 10.1007/s00367-003-0148-8.

Bourry, C., Chazallon, B., Charlou, J.L., Pierre Donval, J., Ruffine, L., Henry, P., et al. (2009). Free gas and gas hydrates from the Sea of Marmara, Turkey. *Chemical Geology* 264(1-4)**,** 197-206. doi: 10.1016/j.chemgeo.2009.03.007.

Charlou, J.L., Donval, J.P., Fouquet, Y., Ondreas, H., Knoery, J., Cochonat, P., et al. (2004). Physical and chemical characterization of gas hydrates and associated methane plumes in the Congo–Angola Basin. *Chemical Geology* 205(3-4)**,** 405-425. doi: 10.1016/j.chemgeo.2003.12.033.

Charlou, J.L., Donval, J.P., Zitter, T., Roy, N., Jean-Baptiste, P., Foucher, J.P., et al. (2003). Evidence of methane venting and geochemistry of brines on mud volcanoes of the eastern Mediterranean Sea. *Deep Sea Research Part I: Oceanographic Research Papers* 50(8)**,** 941-958. doi: 10.1016/s0967-0637(03)00093-1.

Cheng, B., Xu, J., Lu, Z., Li, Y., Wang, W., Yang, S., et al. (2018). Hydrocarbon source for oil and gas indication associated with gas hydrate and its significance in the Qilian Mountain permafrost, Qinghai, Northwest China. *Marine and Petroleum Geology* 89**,** 202-215. doi: 10.1016/j.marpetgeo.2017.02.019.

Choi, J., Kim, J.-H., Torres, M.E., Hong, W.-L., Lee, J.-W., Yi, B.Y., et al. (2013). Gas origin and migration in the Ulleung Basin, East Sea: Results from the Second Ulleung Basin Gas Hydrate Drilling Expedition (UBGH2). *Marine and Petroleum Geology* 47**,** 113-124. doi: 10.1016/j.marpetgeo.2013.05.022.

Claypool, G.E., Milkov, A.V., Lee, Y.J., Torres, M.E., Borowski, W.S., and Tomaru, H., (2006). Microbial methane generation and gas transport in shallow sediments of an accretionary complex, southern Hydrate Ridge (ODP Leg 204), offshore Oregon, USA. *Proceedings of the Ocean Drilling Program: Scientific Results* 204**,** 1-52. doi: <https://doi.org/10.2973/odp.proc.sr.204.113.2006>.

Dai, J., Ni, Y., Huang, S., Peng, W., Han, W., Gong, D., et al. (2017). Genetic types of gas hydrates in China. *Petroleum Exploration and Development* 44(6)**,** 887-898. doi: 10.1016/s1876-3804(17)30101-5.

Dixit, G., Ram, H., and Kumar, P. (2019). Origin of gas in gas hydrates as interpreted from geochemistry data obtained during the National Gas Hydrate Program Expedition 02, Krishna Godavari Basin, offshore India. *Marine and Petroleum Geology* 108**,** 389-396. doi: 10.1016/j.marpetgeo.2018.11.047.

Fang, Y., Wei, J., Lu, H., Liang, J., Lu, J.a., Fu, J., et al. (2019). Chemical and structural characteristics of gas hydrates from the Haima cold seeps in the Qiongdongnan Basin of the South China Sea. *Journal of Asian Earth Sciences* 182. doi: 10.1016/j.jseaes.2019.103924.

Fu, S.Y., and Lu, J.A. (2010). The characteristics and origin of gas hydrate in Shenhu area, South China Sea. *Marine Geology Letters* 26**,** 6-10 (in Chinese with English abstract). doi: 10.16028/j.1009.

Giunta, T., Labidi, J., Kohl, I.E., Ruffine, L., Donval, J.P., Géli, L., et al. (2021). Evidence for methane isotopic bond re-ordering in gas reservoirs sourcing cold seeps from the Sea of Marmara. *Earth and Planetary Science Letters* 553. doi: 10.1016/j.epsl.2020.116619.

Hachikubo, A., Khlystov, O., Krylov, A., Sakagami, H., Minami, H., Nunokawa, Y., et al. (2010a). Molecular and isotopic characteristics of gas hydrate-bound hydrocarbons in southern and central Lake Baikal. *Geo-Marine Letters* 30(3-4)**,** 321-329. doi: 10.1007/s00367-010-0203-1.

Hachikubo, A., Krylov, A., Sakagami, H., Minami, H.N., Y., Shoji, H.M., T., and Jin, Y.K.O., A. (2010b). Isotopic composition of gas hydrates in subsurface sediments subsurface sediments from offshore Sakhalin Island, Sea of Okhotsk. *Geo-Marine Letters* 30**,** 313-319. doi: 10.1007/s00367-009-0178-y).

Hachikubo, A., Yanagawa, K., Tomaru, H., Lu, H.L., and Matsumoto, R. (2015). Molecular and Isotopic Composition of Volatiles in Gas Hydrates and in Sediment from the Joetsu Basin, Eastern Margin of the Japan Sea. *Energies* 8(6)**,** 4647-4666. doi: 10.3390/en8064647.

Hovland, M., Svensen, H., Forsberg, C.F., Johansen, H., Fichler, C., Fosså, J.H., et al. (2005). Complex pockmarks with carbonate-ridges off mid-Norway: Products of sediment degassing. *Marine Geology* 218(1-4)**,** 191-206. doi: 10.1016/j.margeo.2005.04.005.

Ijiri A., Inagaki F., Kubo Y., Adhikari R. R., Hattori S., Hoshino T., et al. (2018). Deep-biosphere methane production stimulated by geofluids in the Nankai accretionary complex. *Science Advances* 4**,** eaao4631.

Ketzer, J.M., Augustin, A., Rodrigues, L.F., Oliveira, R., Praeg, D., Pivel, M.A.G., et al. (2018). Gas seeps and gas hydrates in the Amazon deep-sea fan. *Geo-Marine Letters* 38(5)**,** 429-438. doi: 10.1007/s00367-018-0546-6.

Ketzer, M., Praeg, D., Pivel, M.A.G., Augustin, A.H., Rodrigues, L.F., Viana, A.R., et al. (2019). Gas Seeps at the Edge of the Gas Hydrate Stability Zone on Brazil’s Continental Margin. *Geosciences* 9(5)**,** 193. doi: 10.3390/geosciences9050193.

Kida, M., Hachikubo, A., Sakagami, H., Minami, H., Krylov, A., Yamashita, S., et al. (2009). Natural gas hydrates with locally different cage occupancies and hydration numbers in Lake Baikal. *Geochemistry, Geophysics, Geosystems* 10(5)**,** n/a-n/a. doi: 10.1029/2009gc002473.

Kida, M., Jin, Y., Watanabe, M., Konno, Y., Yoneda, J., Egawa, K., et al. (2015). Chemical and crystallographic characterizations of natural gas hydrates recovered from a production test site in the eastern Nankai Trough. *Marine and Petroleum Geology* 66**,** 396-403. doi: 10.1016/j.marpetgeo.2015.02.019.

Kida, M., Jin, Y., Yoneda, J., Oshima, M., Kato, A., Konno, Y., et al. (2019). Crystallographic and geochemical properties of natural gas hydrates accumulated in the National Gas Hydrate Program Expedition 02 drilling sites in the Krishna-Godavari Basin off India. *Marine and Petroleum Geology* 108**,** 471-481. doi: 10.1016/j.marpetgeo.2018.10.012.

Kida, M., Khlystov, O., Zemskaya, T., Takahashi, N., Minami, H., Sakagami, H., et al. (2006). Coexistence of structure I and II gas hydrates in Lake Baikal suggesting gas sources from microbial and thermogenic origin. *Geophysical Research Letters* 33(24). doi: 10.1029/2006gl028296.

Kim, J.-H., Hachikubo, A., Kida, M., Minami, H., Lee, D.-H., Jin, Y.K., et al. (2020). Upwarding gas source and postgenetic processes in the shallow sediments from the ARAON Mounds, Chukchi Sea. *Journal of Natural Gas Science and Engineering* 76. doi: 10.1016/j.jngse.2020.103223.

Kim, J.-H., Park, M.-H., Chun, J.-H., and Lee, J.Y. (2011). Molecular and isotopic signatures in sediments and gas hydrate of the central/southwestern Ulleung Basin: high alkalinity escape fuelled by biogenically sourced methane. *Geo-Marine Letters* 31(1)**,** 37-49. doi: 10.1007/s00367-010-0214-y.

Kim, J.-H., Torres, M.E., Lee, J.Y., Hong, W.-L., Holland, M., Park, M.-H., et al. (2013). Depressurization experiment of pressure cores from the central Ulleung Basin, East Sea: Insights into gas chemistry. *Organic Geochemistry* 62**,** 86-95. doi: 10.1016/j.orggeochem.2013.07.010.

Klapp, S.A., Murshed, M.M., Pape, T., Klein, H., Bohrmann, G., Brewer, P.G., et al. (2010). Mixed gas hydrate structures at the Chapopote Knoll, southern Gulf of Mexico. *Earth and Planetary Science Letters* 299(1-2)**,** 207-217. doi: 10.1016/j.epsl.2010.09.001.

Koch, S., Schroeder, H., Haeckel, M., Berndt, C., Bialas, J., Papenberg, C., et al. (2016). Gas migration through Opouawe Bank at the Hikurangi margin offshore New Zealand. *Geo-Marine Letters* 36(3)**,** 187-196. doi: 10.1007/s00367-016-0441-y.

Lai, H., Fang, Y., Kuang, Z., Ren, J., Liang, J., Lu, J.a., et al. (2021). Geochemistry, origin and accumulation of natural gas hydrates in the Qiongdongnan Basin, South China Sea: Implications from site GMGS5-W08. *Marine and Petroleum Geology* 123. doi: 10.1016/j.marpetgeo.2020.104774.

Lai, H., Qiu, H., Kuang, Z., Ren, J., Fang, Y., Liang, J., et al. (2022). Integrated signatures of secondary microbial gas within gas hydrate reservoirs: A case study in the Shenhu area, northern South China Sea. *Marine and Petroleum Geology* 136. doi: 10.1016/j.marpetgeo.2021.105486.

Lalk, E., Pape, T., Gruen, D.S., Kaul, N., Karolewski, J.S., Bohrmann, G., et al. (2022). Clumped methane isotopologue-based temperature estimates for sources of methane in marine gas hydrates and associated vent gases. *Geochimica et Cosmochimica Acta*. doi: 10.1016/j.gca.2022.04.013.

Li, A., Cai, F., Wu, N., Li, Q., Yan, G., Sun, Y., et al. (2021). Structural controls on widespread methane seeps in the back-arc basin of the Mid-Okinawa Trough. *Ore Geology Reviews* 129. doi: 10.1016/j.oregeorev.2020.103950.

Liang, J., Wang, J.L., Lu, J.A., Kang, D.J., Kuang, Z.G., and Yang, C.Z. (2017). The characteristics of logging response of the gas hydrate formation in Taixinan Basin and its geological significance. *Earth Science Frontiers* 24**,** 32-40.

Liang, Q., Xiao, X., Zhao, J., Zhang, W., Li, Y., Wu, X., et al. (2022). Geochemistry and sources of hydrate-bound gas in the Shenhu area, northern south China sea: Insights from drilling and gas hydrate production tests. *Journal of Petroleum Science and Engineering* 208. doi: 10.1016/j.petrol.2021.109459.

Liu, C., Meng, Q., He, X., Li, C., Ye, Y., Lu, Z., et al. (2015a). Comparison of the characteristics for natural gas hydrate recovered from marine and terrestrial areas in China. *Journal of Geochemical Exploration* 152**,** 67-74. doi: 10.1016/j.gexplo.2015.02.002.

Liu, C., Meng, Q., He, X., Li, C., Ye, Y., Zhang, G., et al. (2015b). Characterization of natural gas hydrate recovered from Pearl River Mouth basin in South China Sea. *Marine and Petroleum Geology* 61**,** 14-21. doi: 10.1016/j.marpetgeo.2014.11.006.

Lorenson, T.D., and Collett, T.S. (2018). National Gas Hydrate Program Expedition 01 offshore India; gas hydrate systems as revealed by hydrocarbon gas geochemistry. *Marine and Petroleum Geology* 92**,** 477-492. doi: 10.1016/j.marpetgeo.2017.11.011.

Lorenson, T.D., Collett, T.S., and Hunter, R.B. (2011). Gas geochemistry of the Mount Elbert Gas Hydrate Stratigraphic Test Well, Alaska North Slope: Implications for gas hydrate exploration in the Arctic. *Marine and Petroleum Geology* 28(2)**,** 343-360. doi: 10.1016/j.marpetgeo.2010.02.007.

Lorenson, T.D., Whiticar, M.J., Waseda, A., Dallimore, S.R., and Collett, T.S. (1999). "Gas composition and isotopic geochemistry of cuttings, core, and gas hydrate from the JAPEX-JNOC-GSC Mallik 2L-38 gas hydrate research well.," in *Geological Survey of Canada, Bulletin* eds. S.R. Dallimore, T. Uchida & T.S. Collett.), 143-163.

Lorenson, T.D.C., T.S. (2000). "Gas content and composition of gas hydrate from sediments of the southeastern north American continental margin," in *Proceedings of the Ocean Drilling Program, Scientific Results, ,* ed. C.K. Paull, Matsumoto, R., Wallace, P.J., and Dillon, W.P.).

Lu, H., Seo, Y.T., Lee, J.W., Moudrakovski, I., Ripmeester, J.A., Chapman, N.R., et al. (2007). Complex gas hydrate from the Cascadia margin. *Nature* 445(7125)**,** 303-306. doi: 10.1038/nature05463.

Lu, Z., Zhu, Y., Liu, H., Zhang, Y., Jin, C., Huang, X., et al. (2013). Gas source for gas hydrate and its significance in the Qilian Mountain permafrost, Qinghai. *Marine and Petroleum Geology* 43**,** 341-348. doi: 10.1016/j.marpetgeo.2013.01.003.

Lu, Z.Q., Zhu, Y., H., Zhang, Y.Q., Wen, H.J., Li, Y.H., Jia, Z.Y., et al. (2010). Study on genesis of gases from gas hydrate in the Qilian Moutain Permafrost, Qinghai. *Geoscience* 24**,** 581-588.

Lückge, A., Kastner, M., Littke, R., and Cramer, B. (2002). Hydrocarbon gas in the Costa Rica subduction zone: primary composition and post-genetic alteration. *Organic Geochemistry* 33**,** 933-943.

MacDonald, I.R., Bohrmann, G., Escobar, E., Abegg, F., Blanchon, P.B., V., Brückmann, W., et al. (2004). Asphpalt Volcanism and Chemosynthetic life in the Campeche Knolls, Gulf of Mexico. *Science* 304**,** 999-1002.

Mazurenko, L.L., Soloviev, V.A., Belenkaya, I., Ivanov, M.K., and Pinheiro, L.M. (2002). Mud volcano gas hydrates in the Gulf of Cadiz. *Terra Nova* 14(5)**,** 321-329. doi: 10.1046/j.1365-3121.2002.00428.x.

Milkov, A.V., Claypool, G.E., Lee, Y.-J., and Sassen, R. (2005). Gas hydrate systems at Hydrate Ridge offshore Oregon inferred from molecular and isotopic properties of hydrate-bound and void gases. *Geochimica et Cosmochimica Acta* 69(4)**,** 1007-1026. doi: 10.1016/j.gca.2004.08.021.

Miller, D.J., Ketzer, J.M., Viana, A.R., Kowsmann, R.O., Freire, A.F.M., Oreiro, S.G., et al. (2015). Natural gas hydrates in the Rio Grande Cone (Brazil): A new province in the western South Atlantic. *Marine and Petroleum Geology* 67**,** 187-196. doi: 10.1016/j.marpetgeo.2015.05.012.

Pape, T., Bahr, A., Rethemeyer, J., Kessler, J.D., Sahling, H., Hinrichs, K.-U., et al. (2010a). Molecular and isotopic partitioning of low-molecular-weight hydrocarbons during migration and gas hydrate precipitation in deposits of a high-flux seepage site. *Chemical Geology* 269(3-4)**,** 350-363. doi: 10.1016/j.chemgeo.2009.10.009.

Pape, T., Kasten, S., Zabel, M., Bahr, A., Abegg, F., Hohnberg, H.-J., et al. (2010b). Gas hydrates in shallow deposits of the Amsterdam mud volcano, Anaximander Mountains, Northeastern Mediterranean Sea. *Geo-Marine Letters* 30(3-4)**,** 187-206. doi: 10.1007/s00367-010-0197-8.

Pohlman, J.W., Canuel, E.A., Chapman, N.R., Spence, G.D., Whiticar, M.J., and Coffin, R.B. (2005). The origin of thermogenic gas hydrates on the northern Cascadia Margin as inferred from isotopic (13C/12C and D/H) and molecular composition of hydrate and vent gas. *Organic Geochemistry* 36(5)**,** 703-716. doi: 10.1016/j.orggeochem.2005.01.011.

Rodrigues, L., Ketzer, J., Oliveira, R., dos Santos, V., Augustin, A., Cupertino, J., et al. (2019). Molecular and Isotopic Composition of Hydrate-Bound, Dissolved and Free Gases in the Amazon Deep-Sea Fan and Slope Sediments, Brazil. *Geosciences* 9(2)**,** 73. doi: 10.3390/geosciences9020073.

Ruffine, L., Donval, J.-P., Croguennec, C., Burnard, P., Lu, H., Germain, Y., et al. (2018). Multiple gas reservoirs are responsible for the gas emissions along the Marmara fault network. *Deep Sea Research Part II: Topical Studies in Oceanography* 153**,** 48-60. doi: 10.1016/j.dsr2.2017.11.011.

Ryo Matsumoto, T.U., 3 Amane Waseda,3 Tsutomu Uchida,4 Satoshi Takeya,5 Takashi Hirano,6 Kenji Yamada,7 Yuriko Maeda,8 and Tomoharu Okui8 (2000). "Occurrence, structure, and composition of natural gas hydrate recovered from the Blake Ridge Northwest Atlantic," in *Proceedings of the Ocean Drilling Program, Scientific Results, Vol. 164,* ed. C.K. Paull, Matsumoto, R., Wallace, P.J., and Dillon, W.P. .).

Sassen, R., Loshb, S.L.C.I., L., Roberts, H.H., Wheland, J.K., Milkov, A.V., Sweet, S.T., et al. (2001a). Massive vein-filling gas hydrate relation to ongoing gas migration from the deep subsurface in the gulf of mexico. *Marine & Petroleum Geology* 18**,** 551-560.

Sassen, R., Sweet, S.T., Milkov, A.V., DeFreitasI, D.A., Salata, G.G., and McDade, E.C. (1999). Geology and geochemistry of gas hydrates, central gulf of mexico continental slope. *Gulf Coast Association of Geological Societies Transactions* 49**,** 462-468.

Sassen, R., Sweet, S.T., and Milkov, A.V.D.A.D.M.C.K.I. (2001b). Thermogenic vent gas and gas hydrate in the Gulf of Mexico. *Geology* 29**,** 107-110.

Sha, Z.B., Xu, Z.Q., Fu, S.Y., Liang, J.Q., Zhang, W., Su, P.B., et al. (2019). Gas sources and its implications for hydrate accumulation in the eastern Pearl River Mouth Basin. *Marine Geology & Quaternary Geology* 39**,** 116-125.

Snyder, G.T., Sano, Y., Takahata, N., Matsumoto, R., Kakizaki, Y., and Tomaru, H. (2020). Magmatic fluids play a role in the development of active gas chimneys and massive gas hydrates in the Japan Sea. *Chemical Geology* 535. doi: 10.1016/j.chemgeo.2020.119462.

Stadnitskaia, A., Ivanov, M.K., Poludetkina, E.N., Kreulen, R., and van Weering, T.C.E. (2008). Sources of hydrocarbon gases in mud volcanoes from the Sorokin Trough, NE Black Sea, based on molecular and carbon isotopic compositions. *Marine and Petroleum Geology* 25(10)**,** 1040-1057. doi: 10.1016/j.marpetgeo.2007.08.001.

Stern, L.A., and Lorenson, T.D. (2014). Grain-scale imaging and compositional characterization of cryo-preserved India NGHP 01 gas-hydrate-bearing cores. *Marine and Petroleum Geology* 58**,** 206-222. doi: 10.1016/j.marpetgeo.2014.07.027.

Stern, L.A., Lorenson, T.D., and Pinkston, J.C. (2011). Gas hydrate characterization and grain-scale imaging of recovered cores from the Mount Elbert Gas Hydrate Stratigraphic Test Well, Alaska North Slope. *Marine and Petroleum Geology* 28(2)**,** 394-403. doi: 10.1016/j.marpetgeo.2009.08.003.

Vaular, E.N., Barth, T., and Haflidason, H. (2010). The geochemical characteristics of the hydrate-bound gases from the Nyegga pockmark field, Norwegian Sea. *Organic Geochemistry* 41(5)**,** 437-444. doi: 10.1016/j.orggeochem.2010.02.005.

Wang, D.T., Gruen, D.S., Sherwood, L.B., Hinrichs, K.-U., Stewart, L.C., Holden, J.F., et al. (2015a). Nonequilibrium clumped isotope signals in microbial methane. *Science* 348**,** 428-431.

Wang, P., Huang, X., Pang, S., Zhu, Y., Lu, Z., Zhang, S., et al. (2015b). Geochemical dynamics of the gas hydrate system in the Qilian Mountain Permafrost, Qinghai, Northwest China. *Marine and Petroleum Geology* 59**,** 72-90. doi: 10.1016/j.marpetgeo.2014.07.009.

Wei, J., Wu, T., Zhu, L., Fang, Y., Liang, J., Lu, H., et al. (2021). Mixed gas sources induced co-existence of sI and sII gas hydrates in the Qiongdongnan Basin, South China Sea. *Marine and Petroleum Geology* 128. doi: 10.1016/j.marpetgeo.2021.105024.

Winckler, G., Aeschbach-Hertig, W., Holocher, J., Kipfer, R., Levin, I., Poss, C., et al. (2002). Noble gases and radiocarbon in natural gas hydrates. *Geophysical Research Letters* 29(10)**,** 63-61-63-64. doi: 10.1029/2001gl014013.

Xu, C., Wu, N., Sun, Z., Zhang, X., Geng, W., Cao, H., et al. (2021). Assessing methane cycling in the seep sediments of the mid-Okinawa Trough: Insights from pore-water geochemistry and numerical modeling. *Ore Geology Reviews* 129. doi: 10.1016/j.oregeorev.2020.103909.

Ye, J., Qin, X., Qiu, H., Xie, W., Lu, H., Lu, C., et al. (2018). Data Report: Molecular and Isotopic Compositions of the Extracted Gas from China’s First Offshore Natural Gas Hydrate Production Test in South China Sea. *Energies* 11(10)**,** 2793. doi: 10.3390/en11102793.

Ye, J., Wei, J., Liang, J., Lu, J., Lu, H., and Zhang, W. (2019). Complex gas hydrate system in a gas chimney, South China Sea. *Marine and Petroleum Geology* 104**,** 29-39. doi: 10.1016/j.marpetgeo.2019.03.023.

Zhang, N., Snyder, G.T., Lin, M., Nakagawa, M., Gilbert, A., Yoshida, N., et al. (2021). Doubly substituted isotopologues of methane hydrate (13CH3D and 12CH2D2): Implications for methane clumped isotope effects, source apportionments and global hydrate reservoirs. *Geochimica et Cosmochimica Acta* 315**,** 127-151. doi: 10.1016/j.gca.2021.08.027.

Zhang, W., Liang, J.Q., Wei, J.G., Su, P.B., Lin, L., and Huang, W. (2019). Origin of natural gases and associated gas hydrates in the Shenhu area, northern South China Sea: Results from the China gas hydrate drilling expeditions. *J. Asian Earth Sci.* 183**,** 1-16. doi: 10.1016/j.jseaes.2019.103953.