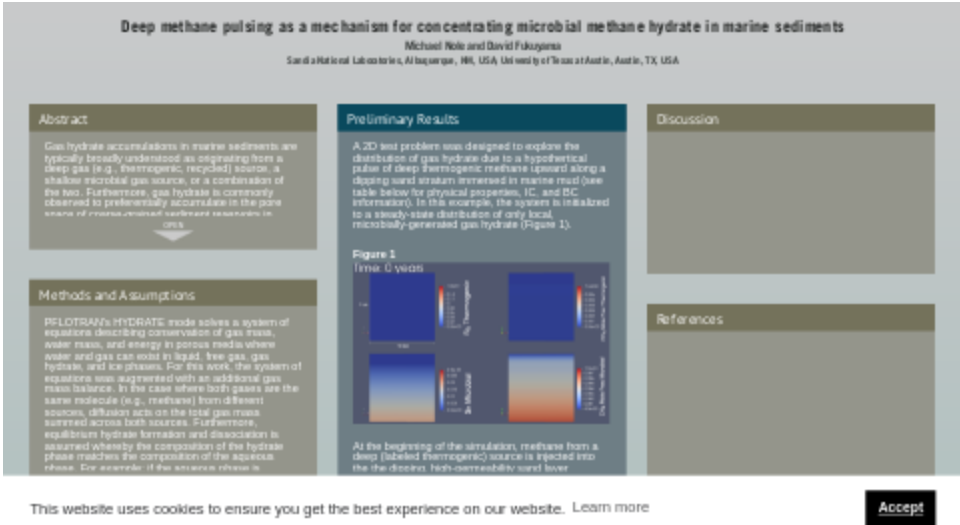


# Deep methane pulsing as a mechanism for concentrating microbial methane hydrate in marine sediments



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## ABSTRACT

Gas hydrate accumulations in marine sediments are typically broadly understood as originating from a deep gas (e.g., thermogenic, recycled) source, a shallow microbial gas source, or a combination of the two. Furthermore, gas hydrate is commonly observed to preferentially accumulate in the pore space of coarse-grained sediment reservoirs in marine environments while remaining largely absent or in a disseminated fracture morphology in surrounding mud sequences. The expression of gas hydrate in nature is therefore controlled by the type of gas source, the gas migration mechanism, and reservoir lithology. The ability to constrain how gas hydrate accumulates is critical for understanding how a gas hydrate reservoir will respond to perturbation, especially in regions of the world that are most sensitive to global climate change. The work presented here describes new development in the open source, high performance subsurface reactive flow and transport simulator PFLOTTRAN to investigate the influence of various methane sources on the expression of gas hydrate in nature. Explicitly tracking methane that is derived from different sources as it moves through the sediment column can provide better estimates of the relative impacts of different gas sources as inputs to global climate models (see Frederick et al. poster B45K-1763 [1] and Eymold et al. poster OS25A-1003 [2]).

## METHODS AND ASSUMPTIONS

PFLOTTRAN's HYDRATE mode solves a system of equations describing conservation of gas mass, water mass, and energy in porous media where water and gas can exist in liquid, free gas, gas hydrate, and ice phases [3] [4] [5]. For this work, the system of equations was augmented with an additional gas mass balance. In the case where both gases are the same molecule (e.g., methane) from different sources, diffusion acts on the total gas mass summed across both sources. Furthermore, equilibrium hydrate formation and dissociation is assumed whereby the composition of the hydrate phase matches the composition of the aqueous phase. For example: if the aqueous phase is composed of 60% methane from Source 1 and 40% from Source 2), the hydrate phase will also be composed of 60% methane from Source 1 and 40% methane from Source 2. For a system where methane from 2 different sources is being tracked, the augmented system of equations looks as follows:

$$\frac{d}{dt} \phi (\sum_i s_i \rho_i x_{m,1}^i) + \nabla \cdot \left( \sum_i (q_i \rho_i x_{m,1}^i - \phi s_i D_i \rho_i \left( \frac{x_{m,1}^i}{x_{m,1}^i + x_{m,2}^i} \right) \nabla (x_{m,1}^i + x_{m,2}^i)) \right) = Q_{m,1}$$

$$\frac{d}{dt} \phi (\sum_i s_i \rho_i x_{m,2}^i) + \nabla \cdot \left( \sum_i (q_i \rho_i x_{m,2}^i - \phi s_i D_i \rho_i \left( \frac{x_{m,2}^i}{x_{m,1}^i + x_{m,2}^i} \right) \nabla (x_{m,1}^i + x_{m,2}^i)) \right) = Q_{m,2}$$

$$\frac{d}{dt} \phi (\sum_i s_i \rho_i x_w^i) + \nabla \cdot (\sum_i (q_i \rho_i x_w^i - \phi s_i D_i \rho_i \nabla x_w^i)) = Q_w$$

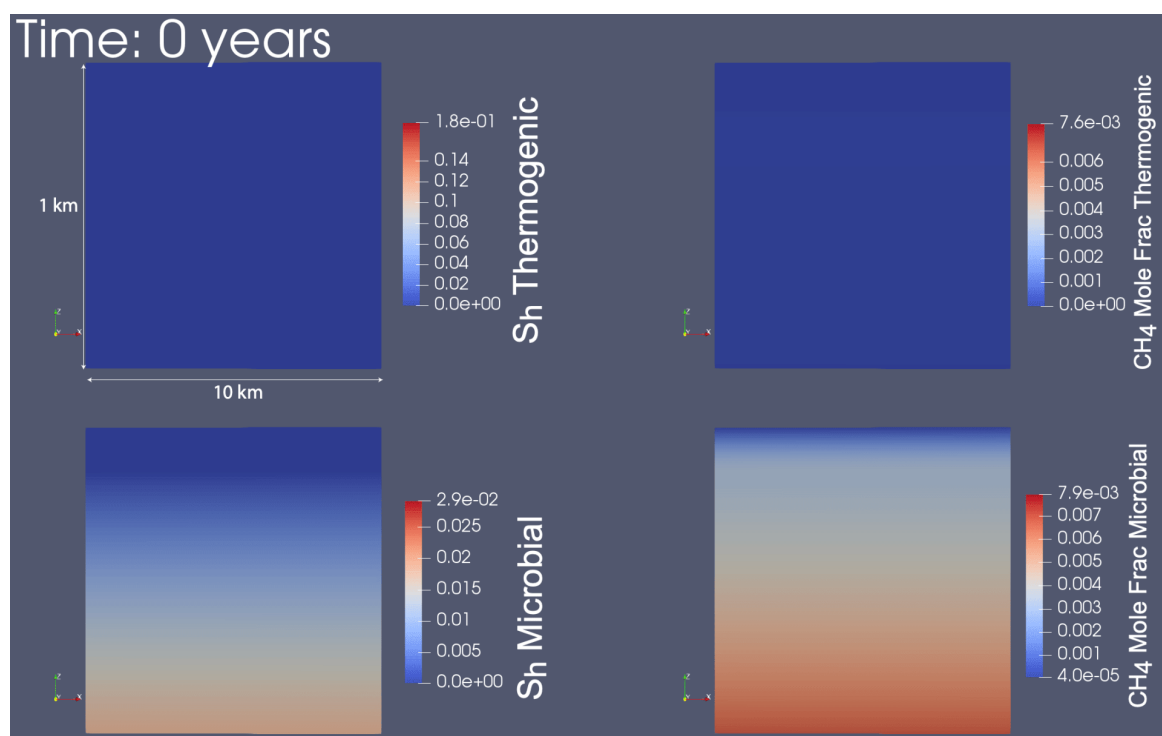
$$\frac{d}{dt} ((1 - \phi) \rho_R C_p T + \sum_i \phi s_i \rho_i U_i) + \sum_i \nabla \cdot (q_i \rho_i H_i) - \nabla \cdot (k \nabla T) = Q_e$$

$$i \in \{L, G, H, I\}$$

## PRELIMINARY RESULTS

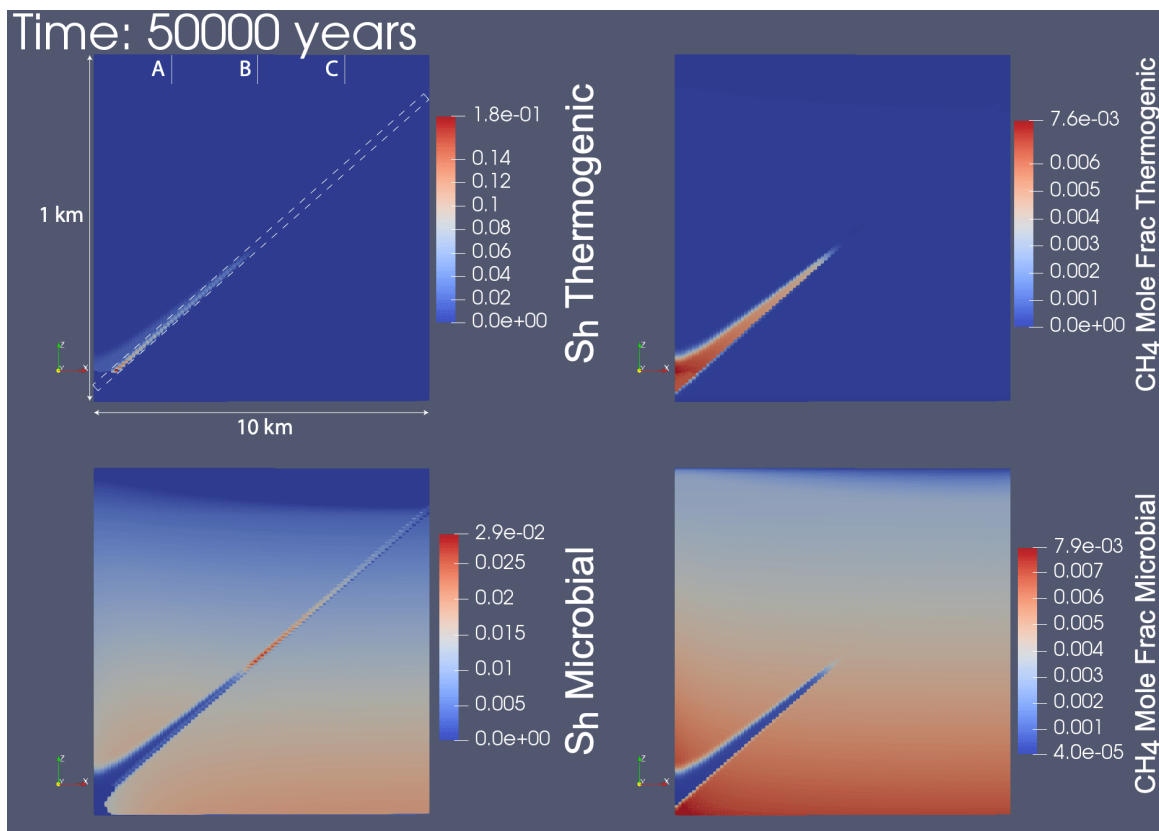
A 2D test problem was designed to explore the distribution of gas hydrate due to a hypothetical pulse of deep thermogenic methane upward along a dipping sand stratum immersed in marine mud (see table below for physical properties, IC, and BC information). In this example, the system is initialized to a steady-state distribution of only local, microbially-generated gas hydrate (Figure 1).

**Figure 1**



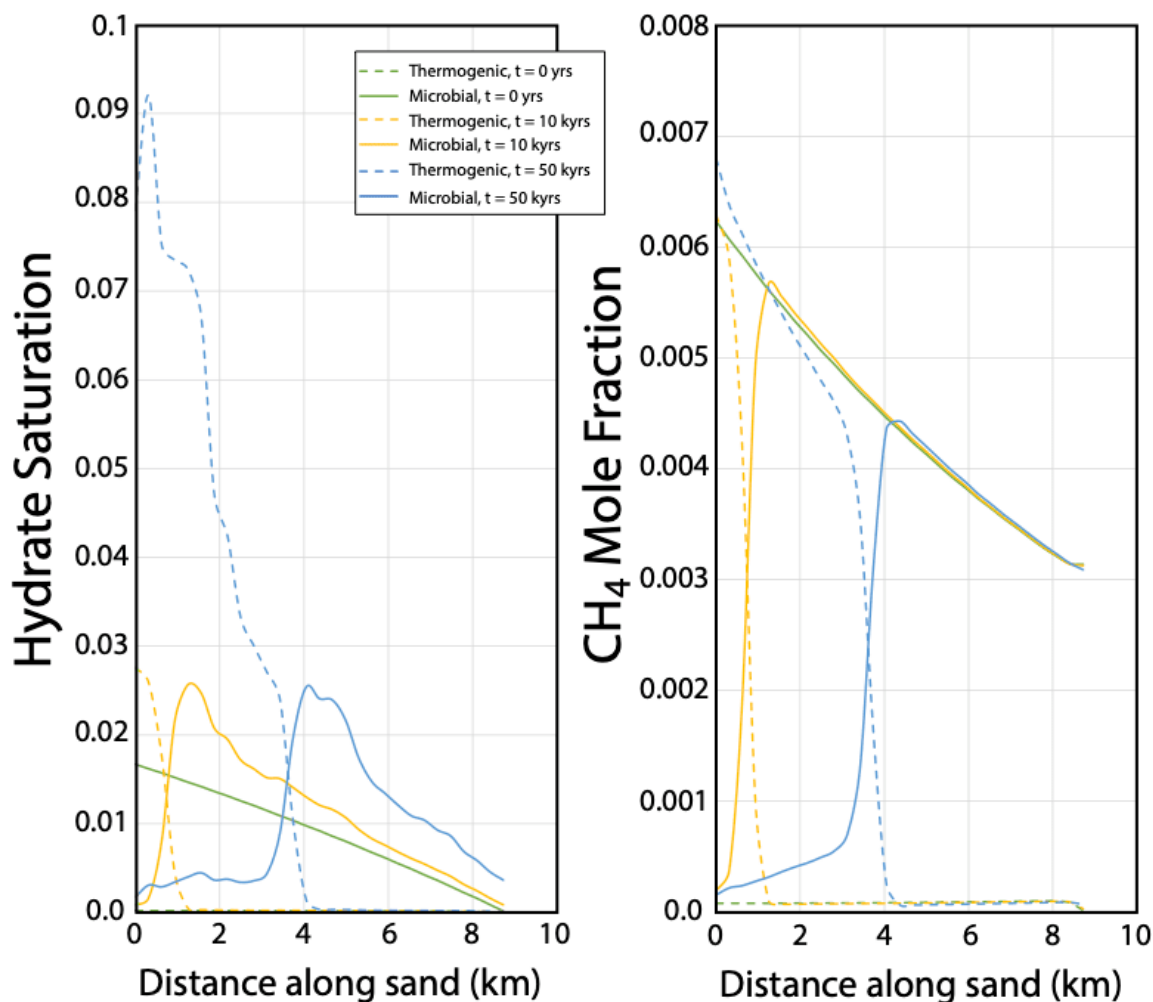
At the beginning of the simulation, methane from a deep (labeled thermogenic) source is injected into the the dipping, high-permeability sand layer (outlined in the dashed line in Figure 2). The system is flushed with deep-sourced methane, which over time becomes the dominant hydrate former at depth (Figure 2). Labels A, B, and C in Figure 2 represent vertical cross-sections in Figure 4.

**Figure 2**



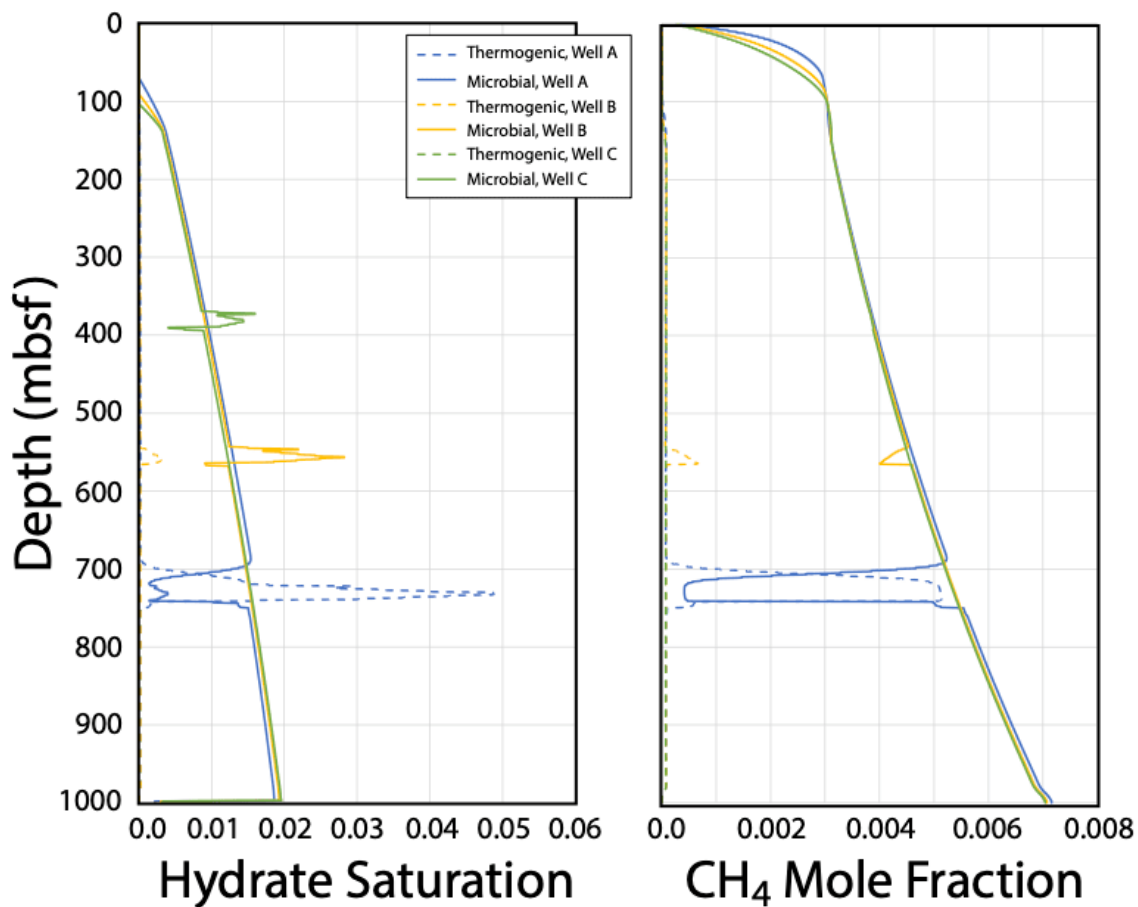
We examine the relative proportions of deep (thermogenic) and shallow (microbial) methane along the dipping sand layer at 3 different times (Figure 3). The down-dip end of the sand corresponds to a distance of 0 in Figure 3. Initially (green line), all dissolved methane and gas hydrate are of shallow microbial origin. Over time, the dissolved methane signature becomes increasingly of deep (thermogenic) origin. Correspondingly, the gas hydrate signature becomes increasingly of deep origin and shallow-origin methane generates higher saturations at shallower depths because it is being pushed upward by the deep methane flux.

**Figure 3**



Three hypothetical vertical transects (wells) are illustrated in Figure 4. If an exploratory Well A were drilled at the end of this simulation, gas hydrate would be found in abundance in the sand layer and would have a predominantly deep-origin (thermogenic) signature. Similarly, dissolved methane would be relatively depleted in shallow-derived methane and enhanced in deep-derived methane. At Well B, gas hydrate would have a predominantly microbial origin but the dissolved methane would show a relative depletion of shallow-derived methane. At Well C, gas hydrate in the sand layer would have an entirely shallow-origin isotopic signature, even though it was concentrated there by a deep methane pulse.

**Figure 4**



Parameter	Microbial Simulation	Thermogenic Simulation
Seafloor Pressure (MPa)	20	20
Seafloor Temperature (°C)	3	3
Geothermal Gradient (°C/km)	19	19
Updip Fluid Flux (mm/yr)	N/A	30
Thermogenic methane concentration in injected fluid (mol/mol)	N/A	$6.5 \times 10^{-3}$
Sand Layer Thickness (m)	N/A	20
Sand Layer Dip Angle (degrees)	N/A	5
Intrinsic Sand x-Permeability (m <sup>2</sup> )	N/A	$1 \times 10^{-13}$
Intrinsic Sand z-Permeability (m <sup>2</sup> )	N/A	$1 \times 10^{-14}$
Intrinsic Mud x-Permeability (m <sup>2</sup> )	$1 \times 10^{-16}$	$1 \times 10^{-16}$
Intrinsic Mud z-Permeability (m <sup>2</sup> )	$1 \times 10^{-17}$	$1 \times 10^{-17}$
Sand Porosity	0.4	0.4
Mud Porosity	0.5	0.5
Initial thermogenic methane concentration (mol/mol)	$1 \times 10^{-5}$	varying
Initial microbial methane concentration (mol/mol)	$1 \times 10^{-5}$	varying
Sedimentation rate (mm/yr)	1	N/A
Seafloor labile organic carbon (%)	1	N/A
Methanogenesis Reaction Rate (s <sup>-1</sup> )	$1 \times 10^{-13}$	N/A

## DISCUSSION

Shallow microbial methane sourcing is generally understood to produce diffuse accumulations of gas hydrate with limited potential to concentrate hydrate in coarse-grained layers over diffusion time scales. Advection of methane from a deep source is often proposed as a preferential accumulation mechanism that can act on shorter timescales. When these two migration mechanisms interact, however, it is unclear what the resulting methane geochemical signature would be, which can make it difficult to characterize a system based off of geochemical data. The results presented here offer insight into simple mixing behavior between deep and shallow-sourced methane at thermodynamic equilibrium.

This type of model could be used not only to constrain the source of methane to a gas hydrate system when given a set of geochemical data (which could provide insight into larger-scale gas migration processes), but it can also be used to interrogate the transport of methane from where it is generated to where it may ultimately be released into the water column or the atmosphere.



## REFERENCES AND ACKNOWLEDGEMENTS

[1] Eymold, W. K., J. M. Frederick, M. A. Nole, B. J. Phrampus and W. T. Wood (2021), Efficient Predictions of Global Free Gas and Gas Hydrate Formation Using K-means Clustering, Abstract OS25A-1003, presented at the 2020 Fall Meeting, AGU, (virtual), 13-17 December.

[2] Frederick, J.M., W.K. Eymold, M. A. Nole, E.W. Conley, B. M. Wagman (2021), Quantifying the Known Unknown: Including Marine Sources of Greenhouse Gases in Climate Modeling, Abstract B45K-1763, presented at the 2020 Fall Meeting, AGU, (virtual), 13-17 December.

[3] Eymold, W. K., J. M. Frederick, M. Nole, B. J. Phrampus and W. T. Wood (2021), Prediction of gas hydrate formation at Blake Ridge using machine learning and probabilistic reservoir simulation, *Geochemistry, Geophysics, Geosystems*, Vol. 22 (4), doi:10.1029/2020GC009574.

[4] Frederick, J. M., W. K. Eymold, M. A. Nole, B. J. Phrampus, T. Lee, W. T. Wood, D. Fukuyama, O. Carty, H. Daigle, H. Yoon, and E. Conley (2021), Forecasting Marine Sediment Properties with Geospatial Machine Learning, Sandia National Laboratories Technical Report, SAND2021-10675.

[5] White, M. D., Kneafsey, T. J., Seol, Y., Waite, W. F., Uchida, S., Lin, J. S., ... & Zyrianova, M. (2020). An international code comparison study on coupled thermal, hydrologic and geomechanical processes of natural gas hydrate-bearing sediments. *Marine and Petroleum Geology*, 104566.

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## ABSTRACT

Naturally occurring gas hydrate accumulations in marine sediments are typically broadly characterized as originating from either thermogenic or microbial methane sources. Gas sources and hydrate reservoirs are linked through corresponding migration mechanisms; often, analyses of gas sources are used to infer migration mechanisms, or vice versa. Gas hydrate is commonly observed to preferentially accumulate in the pore space of coarse-grained sediment strata in marine environments while remaining largely absent or in a disseminated fracture morphology in surrounding mud sequences. This preferential accumulation phenomenon is typically attributed to either preferential flow in coarse-grained strata due to their higher permeability than muds (advective mechanism) or to thermodynamic inhibition to formation in muds because of their small pore size (diffusive mechanism). Accumulation of gas hydrate from a deep thermogenic source originating below the gas hydrate stability zone is often associated with an advective migration mechanism, while concentrated hydrate deposits from locally sourced microbial methane are usually explained with either a diffusive or recycling mechanism. This study presents the development of a multiphase flow and reactive transport simulator, PFLOTRAN, to explicitly track methane that is derived from microbial sources and from deep thermogenic sources. Simulations demonstrate that a pulse of methane-rich fluid of thermogenic origin updip along a high permeability sediment layer can result in a significant region of concentrated hydrate with a predominantly microbial signature. This finding is important to consider when inferring the nature and distribution of a gas hydrate system from observational data.

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