The impact of lithologic heterogeneity and focused fluid flow upon gas hydrate distribution in marine sediments

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Abstract Gas hydrate and free gas accumulation in heterogeneous marine sediment is simulated using a two-dimensional (2-D) numerical model that accounts for mass transfer over geological timescales. The model extends a previously documented one-dimensional (1-D) model such that lateral variations in permeability (k) become important. Various simulations quantitatively demonstrate how focused fluid flow through high-permeability zones affects local hydrate accumulation and saturation. Simulations that approximate a vertical fracture network isolated in a lower permeability shale (kfracture >> kshale) show that focused fluid flow through the gas hydrate stability zone (GHSZ) produces higher saturations of gas hydrate (25–70%) and free gas (30–60%) within the fracture network compared to surrounding shale. Simulations with a dipping, high-permeability sand layer also result in elevated saturations of gas hydrate (60%) and free gas (40%) within the sand because of focused fluid flow through the GHSZ. Increased fluid flux, a deep methane source, or both together increase the effect of flow focusing upon hydrate and free gas distribution and enhance hydrate and free gas concentrations along the high-permeability zones. Permeability anisotropy, with a vertical to horizontal permeability ratio on the order of 102, enhances transport of methane-charged fluid to high-permeability conduits. As a result, gas hydrate concentrations are enhanced within these high-permeability zones. The dip angle of these high-permeability structures affects hydrate distribution because the vertical component of fluid flux dominates focusing effects. Hydrate and free gas saturations can be characterized by a local Peclet number (localized, vertical, focused, and advective flux relative to diffusion) relative to the methane solubility gradient, somewhat analogous to such characterization in 1-D systems. Even in lithologically complex systems, local hydrate and free gas saturations might be characterized by basic parameters (local flux and diffusivity).

1. Introduction

Solid gas hydrates can form when low molecular weight gas molecules and water combine at relatively high pressures, low temperatures, and low-salinity conditions [Sloan and Koh, 2007]. Such conditions occur in sediments along many continental slopes where hydrocarbon gases, especially CH4, accumulate within a depth interval commonly referred to as the gas hydrate stability zone (GHSZ) [Kvenvolden, 1993; Dickens, 2001]. Although the global amount and distribution of marine gas hydrates remain uncertain [e.g., Kvenvolden, 1988; Dickens, 2001, 2011; Milkov, 2004; Archer, 2007; Boswell and Collett, 2011; Burwicz et al., 2011], they may constitute a potential energy resource [Collett, 2002; Walsh et al., 2009], a submarine geohazard [Briaud and Chaouch, 1997; Kwon et al., 2010], and an important constituent of the global carbon cycle [Dickens, 2003, 2011; Archer et al., 2009].

Numerous papers have described gas hydrate abundance across a volumetric region of the seafloor or down a drill site. Recent examples include works regarding the northern Gulf of Mexico [Cook et al., 2008; Boswell et al., 2009, 2012a, 2012b; Lee and Collett, 2012], Krishna-Godavari Basin (offshore eastern India) [Lee and Collett, 2009; Riedel et al., 2010; Wang et al., 2013], Ulleung Basin (offshore South Korea) [Ryu et al., 2009; Kim et al., 2011; Bahk et al., 2013a, 2013b; Lee et al., 2013], and the Shenhu Area (offshore southeast China) [Wang et al., 2010, 2011, 2014]. A common theme among these, and previous studies elsewhere [e.g., Tréhu et al., 2004a; Weinberger et al., 2005; Weinberger and Brown, 2006; Malinverno et al., 2008], is the highly
heterogeneous nature of gas hydrate (Figure 1). Often, at a given location, there exists a first-order vertical profile of gas hydrate saturation (here defined as the fraction of sediment pore space filled with gas hydrate) that spans a lower portion of the GHSZ. Superimposed on this “background” distribution, however, are centimeter-to-meter-scale depth intervals with much higher gas hydrate amounts. Even on outer Blake Ridge, which probably has the simplest stratigraphy of hydrate-bearing locations drilled to date, gas hydrate contents can vary significantly (>4% of pore volume) within a meter [Egeberg and Dickens, 1999; Paull and

Figure 1. (a) Superimposed images of selected LWD data from JIP Leg II at Walker Ridge Sites 313G and H wells on seismic data [Boswell et al., 2012a]. Used with permission. Data show significant deposits of gas hydrates in fractured-filled strata and pore-filled in permeable, coarse-grained sands. (b) Schematic shows four panels (i–iv) to illustrate marine gas hydrate systems: (i) Relationships that exist between three phases of CH₄ in equilibrium, geotherm, hydrotherm, and the finite zone of gas hydrate stability (GHSZ); (ii) a vertical profile of gas hydrate saturation (S_h) in marine sediment within the GHSZ, SO₄²⁻/CO₃ reduction zone, the three-phase boundary, and generic transient and steady state dissolved gas concentration profiles; (iii) a snapshot view of steady state hydrate systems showing the gas hydrate layer overlying a free gas zone that are in equilibrium over geologic timescales and the different sediment and fluid fluxes that interplay due to sedimentation, compaction, and subsidence. The homogeneous lithology causes the hydrate profiles to be uniform in the horizontal dimension; (iv) heterogeneous lithology impacts the hydrate distribution (as evident in Figure 1a) and results in increased saturations in fracture-filled stratigraphy and permeable sand layers as compared to low saturations in the neighboring fine-grained sediment.
The horizons of higher gas hydrate saturation typically are associated with sediment fractures, such as faults, or sediment intervals of greater porosity, such as sand or silt (above references).

To explain gas hydrate accumulation in marine sediment sequences, a series of one-dimensional (1-D) numerical models have been developed and refined [e.g., Rempel and Buffett, 1997; Egeberg and Dickens, 1999; Xu and Ruppel, 1999; Davie and Buffett, 2001; Luff and Wallmann, 2003; Torres et al., 2004; Wallmann et al., 2006; Liu and Fleming, 2007; Bhatnagar et al., 2007; Malinverno et al., 2008]. Although the models differ in details and applicability, they have similar conceptual frameworks: equations for mass, momentum, and energy transport are coupled to understand methane transport, gas hydrate formation, and free gas accumulations in a sediment column at steady state conditions. Simulations resulting from these 1-D models provide a first-order distribution of gas hydrate saturation (and in some cases free gas) at a theoretical borehole. By using site-specific parameters (e.g., water depth, geotherm, sedimentation rate, and organic carbon supply) and including equations for certain dissolved species (e.g., chloride, sulfate, and bicarbonate), the 1-D models can be compared to data collected from actual drill sites [e.g., above references; Bhatnagar et al., 2011; Marquardt et al., 2010; Chatterjee et al., 2011a; Malinverno and Pohlman, 2011]. Ideally, once calibrated, such models might then be used to predict gashydrate distribution in a variety of settings, from large regions of the seafloor to across the globe [Burwicz et al., 2011; Archer et al., 2012; Piñero et al., 2013].

Current 1-D models can explain multiple observations at numerous sites. For example, they simultaneously can explain the first-order distribution of gas hydrate, as well as pore water sulfate and alkalinity profiles [Chatterjee et al., 2011a; Malinverno and Pohlman, 2011]. They also can explain small-scale differences in gas hydrate amounts through differences in sediment porosity [Malinverno and Pohlman, 2011; Cook and Malinverno, 2013]. However, two problems have emerged. First, various 1-D models yield different results at the same location. A good case: the model by Wallmann et al. [2006] renders 0–5 gCH₄/cm² of seafloor for sites on outer Blake Ridge [Marquardt et al., 2010], whereas that by Bhatnagar et al. [2007] gives about 30–40 gCH₄/cm² of seafloor. The latter values are close to those determined through drilling [Dickens et al., 1997; Egeberg and Dickens, 1999]. A consequence of this discrepancy manifests in global gas hydrate estimates and in questions regarding the validity of current modeling. Recent papers [Burwicz et al., 2011; Piñero et al., 2013], which incorporate the Wallmann et al. [2006] model, predict little to no gas hydrate across large areas of continental slopes where abundant gas hydrate likely exists (e.g., Blake Ridge, Cascadia Margin, Gulf of Mexico). Secondly, the 1-D models cannot explain certain heterogeneities in gas hydrate distribution (Figure 1). In particular, fracture networks and dipping permeable layers may be common within continental slope sediment sequences hosting gas hydrate; these features might localize fluid flow, and result in differential accumulation of gas hydrate [Weinberger and Brown, 2006; Tréhu et al., 2006; Riedel et al., 2010; Boswell et al., 2009, 2012a; Frye et al., 2012; Shedd et al., 2012; Sun et al., 2012; Bahk et al., 2013a].

We begin with an assumption: current 1-D models provide the appropriate framework for understanding gas hydrate accumulation, because they explain multiple independent observations at multiple locations. In this paper, we extend one of these 1-D models into two dimensions to understand why heterogeneities might arise from a mechanistic perspective. The new two-dimensional (2-D) model incorporates sedimentation and fluid flow over geological time, and tracks the spatial and temporal accumulation of gas hydrate; it allows us to assess the impact of geological heterogeneities (e.g., interbedding and fractures) and lateral fluid migration upon the concentration and distribution of gas hydrates in marine sediment. Our numerical simulations demonstrate how lithological differences lead to sediment horizons with focused fluid flux, and higher saturations of gas hydrate and free gas.

2. Background

2.1. Observations

Over the last two decades, scientific expeditions have drilled scores of boreholes into continental slope sediment sequences to understand the amount and distribution of gas hydrate below the seafloor. Prior to recent efforts noted above, there were Ocean Drilling Program Leg 146 offshore the Cascadia Margin [e.g., Westbrook et al., 1994], Leg 164 offshore South Carolina [Paull et al., 1996, 2000], Site 1230 on Leg 201 offshore Peru [D’Hondt et al., 2003], Leg 204 offshore Oregon [Tréhu et al., 2003, 2004a], and Integrated
Ocean Drilling Program (IODP) Expedition 311 [Riedel et al., 2006]. At most sites, gas hydrate abundance has been quantified using a variety of independent techniques. Such techniques include measurements on discrete volumes of sediment (e.g., dilution of pore fluids or gas release from pressurized cores), the scanning of sediment cores (e.g., thermal imaging), and well log profiles (e.g., downhole imaging). Good descriptions of these techniques can be found in expedition reports (above) or in review papers [e.g., Tréhu et al., 2006].

Results from analyses at a single borehole often suggest a first-order distribution of gas hydrate abundance with respect to depth (Figure 1). In part because of this finding and in part because of the vertical nature of boreholes, the aforementioned series of 1-D models for gas hydrate accumulation developed. However, at most if not all sites, gas hydrate abundance is more complicated than portrayed through 1-D modeling. For example, at Site 997 (Leg 164) and at Site 1244 (Leg 204), the overall profiles of gas hydrate saturation inferred from resistivity logs and chloride profiles (2–10 %) can be explained nicely through 1-D modeling [e.g., Egeberg and Dickens, 1999; Davie and Buffett, 2001, 2003a; Bhatnagar et al., 2007]. The assumption here is that most gas hydrate exists as very fine grains disseminated in pore space, which is consistent with observations, because almost all cores, when opened, contained no visible gas hydrate. However, at both sites, there were also select meter-scale intervals where large pieces of gas hydrate survived from the time of core retrieval at depth to the time of core opening on the ship. These intervals might be the location of high-angle faults, as perhaps indicated by the shape (vein) and orientation (>30° from normal) of the gas hydrate specimens recovered at Site 1244 [Abegg et al., 2007].

The influence of lithology upon gas hydrate distribution at the meter-scale has been pushed to the forefront with results from recent cruises, two of which we briefly mention here. These are a series of scientific drilling legs in the Krishna-Godavari (KG) Basin (offshore eastern India) conducted by Indian National Gas Hydrate Program (NGHP) and in the northern Gulf of Mexico conducted by the Joint Industry Project (JIP). During the Indian Ocean expedition, a large gas hydrate “deposit” (25% saturated) was discovered at Site NGHP-01-10 across depth interval of ~130 m in fine-grained, fractured (tens of centimeter scale) formation, in addition to other local accumulations of grain-displacing hydrates as nodules or lenses within a large fracture network [Lee and Collett, 2009]. An interesting anomaly confirmed at this site was the absence of prominent, continuous bottom-simulating reflectors (BSRs) which would have otherwise led to disprove the potential abundance of gas hydrates at site NGHP-01-10 [Collett et al., 2008]. These enhanced gas hydrate concentrations in the KG basin are mainly attributed to gas migration from depth through conduits, such as dipping, permeable sediment layers, or faults [Riedel et al., 2010]. The drilling programs in the northern Gulf of Mexico discovered multiple occurrences of saturated, deep-buried, gas hydrates in fracture-dominated, fine-grained pore space, and permeable, interbedded sand sequences [Boswell et al., 2009, 2012a]. These findings confirmed enhanced saturations of gas hydrate occurrence in dipping sand reservoirs (50–90%) above the base of GHSZ (Figure 9a) [Boswell et al., 2012a] and in thick, clay-dominated sequences with fractures [Cook et al., 2008, 2012; Frye et al., 2012]. In all cases, greater gas hydrate abundance has been attributed to a combination of sediment physical properties, localized, vertical fluid flow, and gas flow into the GHSZ through high-flux conduits.

2.2. One-Dimensional Modeling

The series of 1-D numerical models for understanding gas hydrate accumulation in marine sediment [e.g., Rempel and Buffett, 1997; Egeberg and Dickens, 1999; Xu and Ruppel, 1999; Davie and Buffett, 2001, 2003a, 2003b; Gering, 2003; Luff and Wallmann, 2003; Haeckel et al., 2004; Torres et al., 2004; Wallmann et al., 2006; Liu and Flemings, 2006, 2007; Garg et al., 2008] have a similar conceptual framework. Most of these models couple fluid, mass, and energy transport processes with thermodynamics, kinetics, or both to generate methane and to accumulate gas hydrate. The early works were elegant but fairly simple. For example, Davie and Buffett [2001] proposed a 1-D numerical model for hydrate accumulation where methane was supplied from biogenic sources, and gas hydrate precipitated within a prescribed GHSZ upon methane oversaturation. Such modeling, however, was restricted to sites with biogenic methane production and was therefore not applicable to systems with deeper thermogenic sources. Nonetheless, with site-specific observations, such modeling nicely explained first-order observations at several different sites and provided key insights into gas hydrate formation and system behavior.
A drawback of most early numerical models for gas hydrate accumulation is that they do not incorporate biogenic CH4 generated in situ and thermogenic CH4 rising from depth from a generalized perspective. As a consequence, these models are validated only for specific settings containing gas hydrate, most notably Blake Ridge [Egeberg and Dickens, 1999; Davie and Buffett, 2001, 2003a; Gering, 2003; Marquardt et al., 2010] and Cascadia Margin [Luff and Wallmann, 2003; Haeckel et al., 2004; Torres et al., 2004; Liu and Flemings, 2006]. Most previous models have used first-order rate kinetics to model the formation of hydrate in porous media. Moreover, porosity reduction and compaction-driven fluid flow have been modeled using empirical relationships as opposed to using physical and diagenetic processes commonly employed in basin-scale fluid flow models [e.g., Bethke, 1985]. It also means that while the models can explain measured amounts of gas hydrate (i.e., determined using logging, geochemistry, etc.), they often require fine tuning via sensitivity analysis to understand the variations in gas hydrate distribution due to changes in site-specific parameters. In summary, with most models for gas hydrate accumulation, different sites remain disconnected from a mechanistic perspective, in spite of having similar gas hydrate formation and accumulation processes.

Recent modeling efforts have produced a generalized, 1-D, dynamic flow model in thermodynamic equilibrium to quantify the hydrate distribution over geological timescales [Bhatnagar et al., 2007, 2011]. While overcoming some of the issues with other models, this model neglects hydrate formation kinetics. On the other hand, a significant advance of this model is that it relates hydrate occurrence (and underlying processes) at different locations within a unified framework. Furthermore, such modeling uses a scaling scheme to normalize primary variables affecting hydrate saturation on the basis of a few dimensionless groups. Indeed, appropriate scaling of dimensionless groups enables the “collapsing” of a wide range of parameters into simple contour plots to understand basic similarities and differences between gas hydrate abundance at disparate locations. In many cases, two simple plots suffice [Bhatnagar et al., 2007]. One plot summarizes gas hydrate accumulation due to biogenic methane generated within the GHSZ, while the other plot illustrates cases where methane migrates with pore fluids sourced at depth. The modeling establishes a correlation between the net fluid flux and the average hydrate saturation through component balances, thermodynamic equilibrium, and a few key dimensionless groups [Bhatnagar et al., 2007]. The issue is whether such modeling applies and whether similar correlations exist in more complex gas hydrate systems, where variations in lithology and fluid flow become important parameters.

Malinverno [2010] studied natural gas hydrate systems and incorporated heterogeneity into a 1-D model by including thin, horizontal sand layers bound by lower permeability clay layers. This study facilitated the modeling of gas hydrate formation in layered marine sediments by accounting for in situ methane generation and diffusion. As an example, simulations for IODP Site U1325 (Cascadia Margin) predict 30–60% gas hydrate accumulation in pore spaces of thin sand layers (~5 cm thick), but no gas hydrate in thicker (2.5 m), bounding mud layers. Analysis of these simulations show that the diffusive transport of methane from the muds to the sands results in concentrated deposits of hydrate (~50%) in thin sand layers, a finding confirmed in other modeling [Rempel, 2011; Cook and Malinverno, 2013]. This type of modeling, however, cannot explain heterogeneous hydrate formation in the form of nodules and lenses, over larger length scales, or with a significant vertical component.

2.3. Two-Dimensional Modeling

A handful of gas hydrate models have incorporated lithologic heterogeneity and lateral fluid flow to explain local and regional distribution of hydrate occurrence [e.g., Bhatnagar, 2008; Malinverno, 2010; Daigle and Dugan, 2010; Schnürle and Liu, 2011; Frederick and Buffett, 2011]. We briefly note the most recent examples.

Schnürle and Liu [2011] developed a 2-D numerical model to show gas hydrate emplacement in marine sediment. This model is simplistic in the sense that it excludes sedimentation, porosity reduction due to compaction, and lateral variations in temperature, salinity and methane solubility; however, lateral fluid migration and mobile free gas are modeled using a finite-element model that solves for mass, momentum, and energy conservation equations in space and geologic time. Frederick and Buffett [2011] developed a 2-D model showing high gas hydrate saturations in topographic highs, where compaction induces upward fluid flow in sediment with anisotropic permeability.
To fully address gas hydrate distribution in heterogeneous sediments, numerical modeling needs to account for lateral fluid flow and stratigraphic heterogeneity in two or more spatial dimensions. The foundation for such modeling was set up by Bhatnagar [2008], where a generalized 1-D model was extended to two dimensions. Here we benchmark the 2-D model, simulate more complex and heterogeneous gas hydrate settings, and demonstrate similarities between 1-D and 2-D systems. The 2-D model developed and described in this paper is complete as compared to existing dynamic hydrate flow models. The salient features of this model include sedimentation, porosity reduction due to compaction, lateral fluid migration, and mobile free gas coupled with mass and momentum transfer to simulate hydrate distribution and occurrence as a result of fluid flow focusing in heterogeneous lithology.

3. Mathematical Model

3.1. General Framework

The 2-D model presented here spatially extends the 1-D model developed by Bhatnagar et al. [2007], which collectively solves mass balance equations for sediment grains, including solid organic carbon, pore, water, and methane. Labile organic carbon lands on the seafloor and moves down with sedimentation. Sedimentation is associated with compaction, which decreases porosity with increasing depth. Fluids are expelled from pore space as a result of this compaction and move down relative to the seafloor. Upward migration happens when overpressure occurs at depth. Buried organic carbon forms methane via microbially mediated reactions, although the specifics are not detailed. Methane dissolved methane and other pore water species can diffuse through pore space as well as advect with surrounding fluid. Thus, the 2-D model incorporates deposition and compaction of heterogeneous sediment, in situ CH$_4$ generation, and diffusion and advection of dissolved gas. Salinity throughout the sediment column is assumed to be constant and equal to that of the seafloor (~35), although we recognize this induces a minor conceptual problem [Egeberg and Dickens, 1999; Bhatnagar et al., 2007]. Two sediment heterogeneities are considered: (1) high-permeability fracture networks, and (2) high-permeability, interbedded, dipping sand layers.

3.2. Component Mass Balances

The 2-D modeling is formulated with a series of coupled mass balance equations for water, sediment, organic carbon, and methane. The mass balance equations are [Bhatnagar, 2008; Chatterjee et al., 2011b] as follows:

For water,

\[
\frac{\partial}{\partial t} \left[ \phi S_w C_w^i \rho_w + \phi S_{m_w} C_{m_w}^i \rho_m \right] + \nabla \cdot \left[ \phi S_w C_w^i \nu_w \rho_w + \phi S_{m_w} C_{m_w}^i \nu_m \rho_m \right] = 0
\]  

(1)

For sediment,

\[
\frac{\partial}{\partial t} \left[ (1 - \phi) \rho_s + \phi \rho_m \right] + \nabla \cdot \left[ (1 - \phi) \nu_s \rho_s + \phi \nu_m \rho_m \right] = 0
\]  

(2)

For organic carbon,

\[
\frac{\partial}{\partial t} \left[ (1 - \phi) \rho_s \rho_{\text{org}} + \phi \rho_m \rho_{\text{org}} \right] + \nabla \cdot \left[ (1 - \phi) \nu_s \rho_s \rho_{\text{org}} + \phi \nu_m \rho_m \rho_{\text{org}} \right] = -\rho_s \lambda (1 - \phi) \alpha
\]  

(3)

For methane,

\[
\frac{\partial}{\partial t} \left[ \phi S_w C_w^i \rho_w + \phi S_{m_w} C_{m_w}^i \rho_m + \phi S_g C_g^i \rho_g \right] + \nabla \cdot \left[ \phi S_w C_w^i \nu_w \rho_w + \phi S_{m_w} C_{m_w}^i \nu_m \rho_m + \phi S_g C_g^i \nu_g \rho_g \right]
\]

\[
= \nabla \cdot \left[ \phi S_w \rho_w D_m \nabla \cdot C_m^i \rho_{\text{org}} \right] + \frac{M_{\text{org}}}{M_{\text{meth}}} \rho_s \lambda (1 - \phi) \alpha
\]  

(4)

These equations are the same as those described by Bhatnagar et al. [2007] for 1-D simulations, although they are solved in two dimensions. The parameters are $C_i^j$ for mass fraction of component $i$ in phase $j$, $S_j$ for saturation of phase $j$, $\nu_j$ for velocity vector of phase $j$, and $\rho_j$ for density of phase $j$. The kinetic rate constant for methanogenesis reaction is represented by $\lambda$, porosity is denoted by $\phi$, $D_m$ is the diffusivity of methane in seawater and $\alpha$ is the organic carbon content in the sediment. Subscript $s$ corresponds to sediment phase, $w$ to water, and $m$ to methane components. Superscript $l$ corresponds to liquid (water), $h$ to hydrate and $g$ to free gas phases.
3.2. Constitutive Relationships

3.2.1. Water and Free Gas Flux

Our 2-D model accommodates non-hydrostatic conditions and tracks pore pressure as a function of space and time. To do this, water and gas fluxes in a compacting medium are accounted through Darcy’s law [Bear, 1988]:

\[ S_w \phi (v_w - v_s) = \frac{k_{rw}}{\mu_w} \cdot (\nabla p_w - p_w g \nabla z) \]  

\[ S_g \phi (v_g - v_s) = \frac{k_{rg}}{\mu_g} \cdot (\nabla p_g - p_g g \nabla z) \]  

where \( v_w, v_g, \) and \( v_s \) denote water, gas, and sediment velocities, respectively; \( k \) is the absolute sediment permeability tensor; \( k_{rw} \) and \( k_{rg} \) are the relative permeabilities of water and gas; \( \mu_w \) and \( \mu_g \) are the viscosities of water and gas; and \( p_w \) and \( p_g \) are the pore water and gas pressures, respectively.

3.2.2. Sediment Flux

Sedimentation and compaction in the model are assumed to be 1-D with no lateral strain. This enables sediment to deform vertically (in the \( z \) direction) but not horizontally (in the \( x \) or \( y \) direction). Sediment accumulates on the seafloor and moves down with a sediment velocity \( (v_s) \) with positive \( z \) being down. Any gas hydrate within the pore space and any free gas below the critical saturation move downward with the sediment at the same velocity. However, above a critical saturation, free gas becomes mobile, and can migrate up as well as laterally. The sedimentation rate at the seafloor can be defined as follows:

\[ \dot{S} = v_s \quad \text{at} \quad \tilde{z} = 0 \]  

The sediment flux is given by

\[ U_s = \dot{S}(1 - \phi_o) \]  

where \( \phi_o \) is the sediment porosity at the seafloor \( \tilde{z} = 0 \).

3.2.3. Absolute Sediment Permeability

The absolute permeability is an intrinsic property of sediments that allows flow of fluids (e.g., gas and water) through pore space. Absolute sediment permeability is defined by a power law function of porosity [Smith, 1971]:

\[ k = k_o \left( \frac{\phi}{\phi_o} \right)^8 \]  

where \( k_o \) is the initial sediment permeability at the seafloor \( \tilde{z} = 0 \). Reduction in absolute sediment permeability due to gas hydrate formation as a pore-filling structure is modeled by [Kleinberg et al., 2003]

\[ k = k_o \left( 1 - S_h^2 + \frac{2(1 - S_h)^2}{\ln(S_h)} \right) \]  

where \( S_h \) is the hydrate saturation within the pore space.

3.2.4. Relative Permeability of Water and Gas

The ability of different fluids to flow in the presence of each other is measured as relative permeability, and the presence of more than one fluid generally inhibits flow. Relative permeability of water and gas in the presence of each other are modeled as [Bear, 1988] follows:

\[ k_{rw} = k_{rw}^o (S_w^o) \]  

\[ k_{rg} = k_{rg}^o (S_g^o) \]

where \( S_w^o \) and \( S_g^o \) are normalized water and gas saturations, \( k_{rw}^o \) and \( k_{rg}^o \) are the end point relative permeabilities of water and gas, and \( S_{wr} \) and \( S_{gr} \) are the residual water and gas saturations. The water, hydrate, and gas phase saturations must equal unity:

\[ S_w + S_h + S_g = 1 \]
3.2.5. Capillary Pressure

Capillary pressure, \( P_c \), is defined as the difference between gas and pore water pressure:

\[
P_c = p_g - p_w
\]  

(14)

The Leverett J-function, \( J(S_w) \), is used to normalize capillary pressure profiles for different lithologies with different porosities and permeabilities as

\[
J(S_w) = \frac{P_{c,o}(S_w)}{\sigma_{gw} \cos \theta_c \sqrt{\frac{k_o}{\phi_o}}}
\]  

(15)

where \( P_{c,o}(S_w) \) is the capillary pressure at reference absolute permeability \( k_o \) and porosity \( \phi_o \); \( \sigma_{gw} \) is the gas-water surface tension, and \( \theta_c \) is the contact angle. Assuming \( \sigma_{gw} \) and \( \theta_c \) as constants, capillary pressure for any \( \phi \) and \( k \) is

\[
P_c(S_w) = J(S_w) \sigma_{gw} \cos \theta_c \sqrt{\frac{\phi}{k}} = \frac{k_o \phi}{k \phi_o}
\]  

(16)

The reference capillary pressure curve is defined by the Brooks-Corey model [Bear, 1988]:

\[
P_{c,o}(S_w) = P_{c,o}(S_w)^{1/n}
\]  

(17)

where \( P_{c,o} \) is the capillary entry pressure at \( \phi_o \) and \( k_o \), and \( n \) is the pore-size distribution index.

3.2.6. Effective Stress and Porosity

Porosity is modeled as a function of effective stress [Rubey and Hubbert, 1959];

\[
\phi = \phi_w + (\phi_o - \phi_w) e^{\frac{\phi_o - \phi_w}{\phi_o - \phi_w}}
\]  

(18)

where \( \phi_w \) is the minimum porosity achieved at depth, \( \alpha_v \) is total vertical stress (which itself depends on porosity), and \( \sigma_\phi \) is a characteristic stress of compaction.

3.2.7. Total Vertical Stress

Total vertical stress is defined as an integral function of porosity and densities as

\[
\sigma_v = g \int (1 - \phi) \rho_s + \phi \rho_w \eta \frac{dz}{v}
\]  

(19)

3.3. Normalized Variables and Key Dimensionless Groups

As a consequence of generating a generic model for gas hydrate systems, the number of free parameters is large. Following previous work [e.g., Bhatnagar et al., 2007, 2008a, 2011; Chatterjee et al., 2011b], we therefore normalize many equations. Such normalization renders the equations dimensionless, reduces the parameter space and enables expression of results with a few dimensionless numbers.

3.3.1. Porosity Parameters

We define three reduced porosity parameters:

\[
\tilde{\phi} = \frac{\phi - \phi_w}{1 - \phi_w} \quad \eta = \frac{\phi_o - \phi_w}{1 - \phi_w} \quad \gamma = \frac{1 - \phi_w}{\phi_w}
\]  

(20)

3.3.2. Peclet Numbers

Peclet numbers are defined by the ratio of advective fluid flux to methane diffusion (\( D_m \)). Peclet number one, \( Pe_1 \), is characterized by fluid flux related to sedimentation and compaction.

\[
Pe_1 = \frac{U_{f,sed} L_t}{D_m}
\]  

(21)

Peclet number two, \( Pe_2 \), is characterized by the external fluid flux from deeper sediment.

\[
Pe_2 = \frac{U_{f,ext} L_t}{D_m}
\]  

(22)

where \( U_{f,sed} \) is the fluid flux due to sedimentation-compaction for hydrostatic pore pressure conditions, \( U_{f,ext} \) is the fluid flux due to external sources from depth, and \( L_t \) is distance from the seafloor to the base of the GHSZ. The vertical fluid flux due to sedimentation and compaction, \( U_{f,sed} \) is [Bhatnagar et al., 2007]

\[
U_{f,sed} = \frac{1 - \phi_o}{1 - \phi_w} S \phi_w
\]  

(23)
3.3.3. Damköhler Number

The Damköhler number is a dimensionless ratio of the methanogenesis reaction to methane diffusion:

$$Da = \frac{\lambda L_t^2}{D_m}$$

(24)

where $\lambda$ is the first-order rate of methanogenesis reaction.

3.3.4. Other Dimensionless Parameters

Overpressure may characterize many marine gas hydrate systems; that is, pore water pressure at depth may exceed that expected from hydrostatic pressure alone [e.g., Flemings et al., 2003]. Natural gas hydrate systems are often characterized by high sedimentation rates and/or low sediment permeability which lead to overpressure conditions. To quantify the relative importance of these two factors, we define a dimensionless group, $N_{sc}$, that compares absolute sediment permeability to the sedimentation rate [Bhatnagar et al., 2008b]

$$N_{sc} = \frac{k_0 \rho_w g}{\mu_w S}$$

(25)

Large values of $N_{sc}$ imply high-sediment permeability and/or low sedimentation rate, which result in pore water pressures close to hydrostatic pressures. Conversely, smaller $N_{sc}$ values imply low permeability and/or high sedimentation rate, thereby producing overpressure. Dimensionless groups similar to $N_{sc}$ have been defined in earlier 1-D compaction models [Yang and Fowler, 1998; Gutierrez and Wangen, 2005].

The ratio of characteristic compaction depth to the depth to the base of the GHSZ is defined as follows:

$$N_{t\phi} = \frac{L'_{\phi}}{L_t} = \frac{\sigma_v / \rho_w g}{L_t}$$

(26)

Note that the new dimensionless group $N_{t\phi}$ is slightly different from $N_{t\phi}$ (defined as $L_t / L_{\phi}$, where $L_{\phi} = (1 - \phi_w) \cdot (\rho_s / \rho_w - 1)$) [Bhatnagar et al., 2007].

3.3.5. Scaled Variables

The model accounts for three CH$_4$ phases: dissolved, gas hydrate, and free gas. The normalized CH$_4$ phase concentrations are:

$$\tilde{c}_m^l = \frac{c_m^l}{c_{m, eqb}} \quad \tilde{c}_m^h = \frac{c_m^h}{c_{m, eqb}} \quad \tilde{c}_m^g = \frac{c_m^g}{c_{m, eqb}}$$

(27)

where $c_{m, eqb}$ is the solubility of methane in seawater at the base of the GHSZ.

Lithostatic stress, water, gas, and capillary pressures are normalized by the hydrostatic pore water pressure at the base of the GHSZ:

$$\tilde{\sigma}_v = \frac{\sigma_v}{\rho_w g L_t} \quad \tilde{p}_w = \frac{p_w}{\rho_w g L_t} \quad \tilde{p}_g = \frac{p_g}{\rho_w g L_t} \quad \tilde{p}_c = \frac{p_c}{\rho_w g L_t}$$

(28)

The vertical depth and lateral distance are scaled to the depth to the base of the GHSZ:

$$\tilde{z} = \frac{z}{L_t} \quad \tilde{x} = \frac{x}{L_t}$$

(29)

The dimensionless time is defined as:

$$\tilde{t} = \frac{t}{L_t^2 / D_m}$$

(30)

All phase densities are scaled by the density of water as:

$$\tilde{\rho}_m = \frac{\rho_m}{\rho_w} \quad \tilde{\rho}_g = \frac{\rho_g}{\rho_w} \quad \tilde{\rho}_c = \frac{\rho_c}{\rho_w}$$

(31)

The sediment velocity $v_s$ is normalized by the sedimentation rate at the seafloor $\dot{S}$.

$$\tilde{v}_s = \frac{v_s}{\dot{S}}$$

(32)

Finally, organic carbon content and initial organic carbon content are scaled as:

$$\tilde{\alpha} = \frac{\alpha}{\alpha_0} \quad \beta = \frac{\alpha_0}{c_{m, eqb}}$$

(33)

where $\alpha_0$ is the organic carbon content at the seafloor.
conditions to obtain transient and steady state solutions over space and geologic time. The four coupled, dimensionless mass balance equations (equations (A2), (A10), (A13), and (A16)) are solved using a fully implicit numerical scheme using the initial conditions and boundary conditions discussed in Appendix A. A single-point, upstream weighting was used to formulate the relative permeability terms. The solubility of methane dissolved in pore water is calculated using rigorous thermodynamic calculations [Bhatnagar et al., 2007]. The model includes a phase switch algorithm with changes the phase of methane from dissolved phase to hydrate or free gas phases in thermodynamic equilibrium when local methane concentrations exceed the solubility. The primary variables are \( p_w \), \( \bar{V}_w \), \( \bar{n} \) and \( \bar{z}_m \). \( S_h \) or \( S_g \) depending on the local thermodynamic conditions of the grid block at any given time. All the component mass balance equations are then recast in their residual form and the Newton-Raphson method is implemented to iterate on them to converge to the finite solution.

4. Two-Dimensional Model Development and Validation

The 2-D model presented here can be tested, validated, and benchmarked against 1-D results [Bhatnagar et al., 2007]. For this exercise, we use the transport and geologic parameters at the “classic” gas hydrate location — Blake Ridge. The seafloor parameters are seafloor depth = 2700 m below sea level, seafloor temperature = 3°C, and geothermal gradient = 0.04°C/m. The primary dimensionless transport parameters defined are \( \phi_e = 0.1, \phi_g = 10, \beta = 6, \gamma = 9, N_{sc} = 10^3, N_{nc} = 1.485 \). Critical gas saturation, \( S_{gr} \), is specified as 100%, so free gas is immobile. We emphasize that this base case simulation represents systems dominated by fluid flux with no free gas migration. The 2-D code was first tested with homogeneous sediment permeability (i.e., permeability varies vertically due to burial and compaction but does not vary laterally), essentially a 1-D implementation of the 2-D model. Steady state hydrate and free gas saturations were simulated (Figure 2). The hydrate saturation with only in situ methane sources in a homogeneous sediment column increases to a peak saturation of ~11% at the base of the GHSZ with an average hydrate saturation equal to 5.7%. The 2-D profiles match the 1-D results (Figure 3).

There is no flow focusing or preferential accumulation of hydrate and free gas within the sediment in the homogeneous system. To assess the importance of deeper methane sources on hydrate and free gas saturations, methane concentration is specified in pore fluid migrating upward from the base of the model domain (\( \bar{2} = 2 \)) for cases simulated with upward fluid flux (\( \phi_g < 0 \)). This enables methane-charged fluids to migrate upward from deeper sources. This increased methane supply results in higher hydrate and free gas saturations. The average hydrate saturation (\( \langle S_h \rangle \)) is computed within the hydrate stability zone, multiplied...
by $P_{e1}$, and this product ($P_{e1} < S_h >$) is related with the net fluid flux ($P_{e1} + P_{e2}$). The 2-D homogeneous results match the net fluid flux to the average hydrate saturation and validated against previous 1-D results (Figure 3). Therefore, such correlations can now be used to quantify average hydrate saturations in homogeneous sediment using net fluid flux as the primary input. Notably, the assumption of negligible volume change due to methane dissolved in water in the 1-D model approximates the mass fraction of water in liquid phase, $c_l = 1$ [Bhatnagar et al., 2007]. In the 2-D model, volume change due to methane is not assumed to be zero and thus results in a small change in liquid volume when methane dissolves in water. Therefore, the volumetric net fluid flux, ($P_{e1} + P_{e2}$) for 2-D homogeneous cases shows a slight deviation from the 1-D results with increasing external flux (Figure 3).

With the validated 2-D model, heterogeneities (vertical fracture systems and high-permeability interbedded layers) are simulated to show how high-permeability conduits localize fluid flux, and how these localized fluxes result in concentrated and heterogeneous hydrate deposits.

5. Results

5.1. Effect of Vertical Fracture Systems

Fractures, of varying length scales, are common to many settings with gas hydrate, such as the Cascadia Margin [Tréhu et al., 2004a; Weinberger and Brown, 2006] or the southeast Indian Margin [Riedel et al., 2010]. In this study, we have quantified the impact of such fractures by implementing a thin vertical zone of higher permeability in a model for gas hydrate accumulation (Figure 4). These vertical fracture systems are modeled from $\iota = 0$ assuming the fractures extend from the seafloor to the base of the model domain. Obviously, this approximation is simplistic, but it allows gas hydrate and free gas accumulation to be tracked over time while providing first-order insights into the role of focused fluid flow. Simulations are performed with a vertical fracture system 100 times more permeable than surrounding sediment (i.e., $N_{sc} = 10^2$ for sediment and $10^4$ for the vertical fracture system).

The simulations with a vertical fracture network show that focused fluid flow results in higher saturations of gas hydrate and free gas within the
Figure 5. Steady state gas hydrate and free gas saturation contours for isotropic system \((k_h/k_i = 1)\) with biogenic in situ source \((P_{E2} = 0)\) and a vertical fracture system. The location of the fracture is shown by a set of white, vertical dashed lines. A vector field plot shown by white arrows represents the focused fluid flow. The fluid flow within the sediment is in the downward direction because it is plotted relative to the seafloor. The effect of the fracture in focusing flow is clearly illustrated through enhanced hydrate and free gas saturations within the high-permeability conduit. The following parameters were used for this simulation: \(P_{E1} = 0.1, P_{E2} = 0, Da = 10, \beta = 6, \zeta_{mext} = 0, \gamma = 6/9, k_h/k_i = 1, N_{hp} = 1.485,\) and \(N_{sc} = 10^2\), those in a 1-D simulation with homogeneous sediment. However, the gas hydrate and free gas saturations in the vicinity of the fracture system are lower than predicted by the 1-D model because the methane-charged water from these regions is focused into the high-permeability fracture system. To summarize, the focusing of fluid flow within a fracture system changes gas hydrate abundance across an area significantly (Figure 5).

A second case is simulated with a specified fluid flux from external sources at depth. This might represent a setting where thermogenic methane arrives from depth such as along tectonically active margins [e.g., Boswell et al., 2012a]. Higher gas hydrate and free gas saturations are predicted in general (as noted by others e.g., Archer et al., 2012) due to increased fluid flux and greater methane input into the system. Our simulations further show gas higher hydrate and free gas saturation within the high-permeability fracture (Figure 6), as compared to a case with only biogenic in situ methane (Figure 5). Peak hydrate and free gas saturations within the fracture are 48% and 42%, respectively, whereas peak saturations are only 11% and 9% in the neighboring low-permeability sediment. However, and similar to our simulation with only a biogenic methane sources (Figure 5), the values are identical to those in 1-D modeling away from the fracture system. In summary, the fluid flux is amplified within the high-permeability conduit because of a greater flux specified at the base of the modeling domain. Notably, fluid flow in the vicinity of the fracture system is nearly horizontal as a result of focusing.

The above results explain how increased fluid flux results in higher gas hydrate and free gas accumulations along high-permeability zone compared to surrounding lower-permeability sediment. In a fracture system with isotropic permeability \((k_h/fraction = k_i/fraction\) and \(k_h/clay = k_i/clay\), steady state, peak hydrate saturation within the fracture system can be ~26%, while that in the surrounding sediment matrix is only ~10% (Figure 5). Steady state free gas abundances also have a peak saturation of 29% within the fracture column (at the base of the domain) but only 10% in the neighboring shale matrix.

The critical gas saturation is assumed 100% to set the free gas immobile. Consequently, free gas peaks at the base of the domain. These simulation cases emphasize the effect of focusing methane-charged liquid (water) within high-permeability zones. Importantly, gas hydrate and free gas saturations away from the fracture system (i.e., in the lower-permeability formation) are identical to...
fractures or networks thereof. At the simplest level, a higher permeability facilitates increased methane flux and thus increased saturations. Adding an external source of methane at depth results in a greater methane throughput, and consequently amplifies the focusing effect, which yields higher gas hydrate saturations.

5.2. Effect of Permeability Anisotropy

Many sediment sequences on continental slopes are anisotropic, where the ratio of vertical to horizontal permeability \( k_v/k_h \) is less than unity, at least over certain depth intervals. Conceptually, greater horizontal permeability might help focus fluids from low-permeability regions to high-permeability conduits; that is, in anisotropic systems with \( k_v/k_h \) ratios on the order of \( 10^{-2} \) or lower, higher gas hydrate saturations might occur within high-permeability conduits, at least compared to isotropic cases. We therefore simulate a third case, one that has two sources of methane (an internal source within the sediment and an external source from below), but where the sediment is anisotropic in two dimensions. These simulations show peak hydrate and gas saturations of 53% and 40%, at the base of GHSZ and the modeling domain, respectively (Figure 7). Increased horizontal permeability results in more fluid flux toward the fracture system, especially including from sediment farther away.

5.3. Local Flux Within High-Permeability Zones

These steady state simulation results are used to evaluate the local fluid flux and local hydrate saturation. First, the steady state fluid fluxes are computed and recorded locally within the high-permeability zones. Second, the hydrate saturations are noted within the high-permeability zones above the GHSZ. In all the above heterogeneous cases with a vertical fracture system, the localized, focused fluid flux can be computed and related to the average hydrate saturation within the high-permeability conduit (Figure 8). Therefore, gas hydrate saturations in these lithologically heterogeneous systems can be characterized by a local Peclet number \( (P_{e_{local}}) \) characterization is comparable with the concept of a net Peclet number \( (P_{e < S_h >} + P_{e_{ext}}) \), which can be used to characterize 1-D homogeneous systems (Figure 3). Apparently, local hydrate and free gas saturations can be characterized by basic parameters (local flux and diffusivity), even in lithologically complex systems.
5.4. Effect of Permeability Contrast

In the three cases discussed above, gas hydrate and free gas saturations are shown to increase when the permeability of fracture is 100 times that of the neighboring shale matrix. As the permeability contrast between a fracture and surrounding clay increases from 2 to 1000, such modeling further documents a general response: greater gas hydrate and free gas accumulation within the high-permeability conduits (Figure 9). This may explain the observed elevated saturations along fractures [e.g., Weinberger and Brown, 2006; Riedel et al., 2010; Boswell et al., 2012a; Bahk et al., 2013a], as well as beneath areas of active seafloor venting in high-flux conduits like pipes, vents, and dipping reservoirs in advective systems.

Three key findings characterize our plots of gas hydrate accumulation in sediment with fractures (Figure 9). First, the peak saturations and the average saturations both show an increasing trend, albeit to a less degree for average values (Figure 9). The peak hydrate saturations at the base of the GHSZ and the peak free gas concentrations at the base of the domain show maximum (or close to maximum) values. However, the average saturation values across space have considerable scope to increase as hydrates can accumulate in the shallow pore space. Second, the three cases discussed above (e.g., isotropic with biogenic source, isotropic biogenic and deep methane sources, and anisotropic cases with both sources) show an increasing trend in saturations with increasing permeability contrast (Figure 9). Clearly, the fluid focusing ability within the high-permeability conduits increases with the permeability contrast. Moreover, there is increased fluid focusing within these conduits as more methane-charged fluids flow as a result of external flux deeper sources and inclusion of anisotropy. Third, the hydrate saturations show considerable increase with anisotropy effects. However, the free gas saturations are not strongly dependent on anisotropy effects. This effect is due to immobility of the free gas phase (vertically and laterally) as set in these simulations.
5.5. Effect of Horizontal, High-Permeability Layers

Heterogeneity can be modeled as a horizontal layer parallel to the seafloor, analogous to Malinverno [2010] but including advection. This horizontal layer is buried, and its downward movement is tracked in geologic time. However, it is acknowledged that a horizontal layer alone would not represent lateral heterogeneity. In addition, two vertical fracture systems are coupled to two ends of the horizontal, high-permeability layer, and the coupled permeability conduit moves down with sedimentation relative to the seafloor (Figure 10). This configuration allows the fluids to migrate from the bottom of the domain to the seafloor (Figure 11). Due to its horizontal configuration, the dip angle (relative to the horizontal) is zero. Likewise, the above vertical fracture simulations represented systems with a dip angle of 90°. These example simulations are completed to provide a clear understanding of the system and to assess the dependence of dip angle of these permeable layers.

Simulations are run with the same parameter set used in Figure 6, while the horizontal layer and vertical fractures are buried down through time. The temporal profiles are tracked as the sand layer moves down during burial. The transient results show high-focused fluid flow through the permeable layers from depth to the horizontal layer, then across the simulation domain, and then toward the seafloor (Figure 11). Unlike the vertical fracture systems extending through the GHSZ, gas hydrate and free gas do not accumulate along the horizontal layer. Even though high-fluid flux gets focused and flows through the permeable conduit, it does not result in elevated hydrate saturation. The hydrate saturation within the horizontal layer is laterally uniform at 12%. The peaks in hydrate saturation occur at the juncture of the horizontal layer and the vertical fracture on either sides (19% and 32%). Therefore, the localized fluid flux within these horizontal systems cannot be used to evaluate local gas hydrate saturation and distribution. Thus, it is determined that these horizontal fluid flux components do not influence hydrate precipitation. Following a detailed mathematical analysis shown in Appendix C, it is shown that the vertical component of the focused fluid flux in heterogeneous lithology relative to the methane solubility gradient is what drives these elevated hydrate saturations in these lithologically complex systems.

5.6. Effect of Dip Angle of High-Permeability Layers

A steady state, 1-D analytical theory is derived to illustrate the effect of dip angle of the high-permeability layer. The mathematical model shows that the
inclination of the permeable conduit (to the horizontal axis) dictates hydrate saturation and distribution (Appendix C). The net fluid flux \((\bar{P}_h < S_h >)\) is related to the average hydrate flux \((\bar{P}_h (S_h))\) within a fluid conduit inclined in any orientation to the horizontal (not shown here). This generates a series of plots for a range of dip angle values relating the net fluid flux and the average hydrate flux. However, this does not generalize results as these plots depend on the dip angle. Therefore, instead of the net fluid flux, we relate the vertical fluid flux \((\bar{P}_v + \bar{P}_s \sin \theta)\) to the average hydrate flux \((\bar{P}_h (S_h))\). This eliminates the dependence of our results on the dip angle, and the model can generalize a wide range of configurations into one single contour plot (Figure 12), thereby reducing the parameter space. These plots can be used to compute the average hydrate saturation given the fluid flux and the methane solubility gradient for marine hydrate systems in any configuration. It is shown that the vertical fluid flux is what dictates hydrate saturation and accumulation as opposed to the net fluid flux for a case where the conduit is inclined at an angle, \(\theta\) to the horizontal. These results can be extended to any geologic setting, by adapting the model to site-specific parameters [Westbrook et al., 1994; Paull et al., 1996; Riedel et al., 2006; Bhatnagar et al., 2007, 2011] (Figure 12).

5.7. Effect of Free Gas Migration

Focusing of liquid (water) enhances hydrate and free gas saturations within the high-permeability zones. However, free gas can also migrate vertically and laterally if the critical gas saturation is exceeded. To simulate such migration, we reduce the critical gas saturation \((S_g)\) from 100% (immobile) to 5%; therefore, free gas exceeding this critical value will migrate according to Darcy’s law (equation (6)). Free gas migrates upward due to buoyancy and gets sealed by the low-permeability hydrate layer at the base of the GHSZ. Hydrate formation at the base of the GHSZ causes an increase in the capillary entry pressure and creates a hindrance for free gas to enter the GHSZ from below. This results in accumulation of free gas beneath the GHSZ into a connected gas column [Tréhu et al., 2004b].

5.7.1. Vertical Fracture Systems

Simulations show free gas accumulation below the base of the GHSZ (Figure 13). A gas column is formed before the gas pressure exceeds the capillary entry pressure, and gas migrates into the GHSZ. Free gas converts to hydrate immediately at the base of the GHSZ giving rise to a spike in hydrate saturation. Some models track pore water salinity changes to predict free gas migration into the GHSZ [Liu and Flemings, 2006, 2007], but our model assumes quasi steady state, and at these timescales, salinity changes can be assumed constant. Hence, phase equilibrium is unaffected even when free gas migrates into the GHSZ. The high hydrate saturation in the fracture zone causes a permeability reduction resulting in lateral fluid migration beneath the GHSZ. High hydrate saturation also restricts further free gas invasion into the GHSZ. Free gas then migrates laterally from the high-permeability fracture system to the neighboring sediment below the

Figure 12. Average hydrate flux, \(\bar{P}_h < S_h >\) is related to the net vertical fluid flux for marine hydrate systems inclined in any angle to the horizontal axis. Systems with horizontal layers correspond to zero, whereas vertical fractures correspond to 90°. As orientation changes from vertical to horizontal, the saturation decreases, even though there is high flux through these systems. This is represented by multiple lines, albeit shorter length as the vertical component of \(\bar{P}_s \sin \theta\) reduces as orientation changes from vertical to horizontal. Note, the longest line for a given set of lines (given site) represent the vertical conduits with dip angle 90°, and results match the blue curve \((\Delta a = 0)\) in Figure 3. This demonstrates that the vertical fluid flux relative to methane solubility gradient is what drives hydrate saturation and accumulation in these marine hydrate systems. Similarly, this can be extended to 2-D systems where localized vertical fluid flux relative to methane solubility gradient can be correlated to localized average hydrate saturation in systems with complex lithology. These results are adapted for site-specific transport and geologic parameters that resemble three distinct hydrate settings (Blake Ridge site 997, Cascadia Margin Sites U1325 and 889).
base of the GHSZ. Peak hydrate and free gas saturations reach 75% and 62% right above and below the base of the GHSZ within the fracture system. At this enhanced saturation, free gas is mobile and migrates laterally, causing sediment near the fracture to also have relatively higher saturations.

The length of a connected gas column has been shown to be dependent on the dimensionless group $N_{sc}$ [Bhatnagar et al., 2008b]. Lower values of $N_{sc}$ (lower permeability and/or higher sedimentation rate) result in development of higher overpressure in the system, which further results in thicker gas columns and increased gas pressure at the base of the GHSZ close to the lithostatic stress. Fractures tend to open up at this point [Hornbach et al., 2004; Daigle and Dugan, 2010]. This fracturing process is not modeled in any of these simulations, and the simulations are terminated as soon as gas pressure equals total vertical stress.

5.7.2. Dipping Sand Layers

Lithologic heterogeneity is modeled in another configuration (non-vertical), in which dipping sand beds are isolated by low-permeability clay. To model a sand layer, a high-permeability layer is included at a given dip angle (~3°) within the sediment (Figure 14) and buried through geologic time. Sediment compacts with burial. The downward movement of this sand layer and the corresponding transient hydrate and free gas evolution are recorded. The sand layer is assigned an absolute permeability 100 times greater than the surrounding anisotropic shale ($k_v/k_h = 10^{-2}$ for shales and $k_v/k_h = 1$ for sand). The seafloor and transport parameters are same as in the simulations with vertical fracture network except $Da = 1$. During burial, absolute permeability of any grid block can be computed by interpolation, and therefore, interface between zones of different permeabilities are recorded over time. The interface position is used to track the location of the sediment over time.

Systems with dipping sand layers show localized, enhanced concentrations of hydrate and free gas within the high-permeability sand layers (Figure 15). Free gas is focused within the sand layer, and a portion of that flow is in the vertical direction. Peak hydrate saturation within the sand layer is about 59%, higher than the 43% peak hydrate saturation in the low-permeability sediment within the GHSZ. Peak free gas saturation is 38% within the sand layer just below the GHSZ, and similar to fracture network case (Figure 13), free gas migrates laterally into the low-permeability sediment around the sand layer. Thus, the presence of higher-permeability sand layer leads to relatively higher fluid focusing and hydrate saturation within the GHSZ. These results elucidate that lithology alone does not play a significant role in accumulating gas hydrate and free gas but both lithology and structure are important factors for heterogeneous accumulations of gas hydrate and free gas.
lateral methane solubility gradient is zero at any depth. In this and previous work, it is shown that the excess methane concentration beyond the solubility values accumulate as hydrate and free gas in thermodynamic equilibrium. Due to invariant methane solubility in the lateral dimension, there is no excess methane accumulation in the horizontal layers regardless of the fluid focusing effects. However, the hydrates tend to accumulate in vertical or dipping conduits (with considerable dip angle) due to the influence of the vertical component of the flow and the vertical gradient in methane solubility. Third, the local vertical fluid flux within these high-permeability zones can be quantified and related to the local average hydrate saturations. This forms a premise for evaluating hydrate and free gas saturation with the help of the vertical fluid flux as a direct proxy. This work summarizes the basis of fluid focusing effects and quantifies the amount and distribution of hydrates in marine sediment dominated by heterogeneous and complex lithology and stratigraphy.

Some of the model applications include assessing complex, heterogeneous 2-D gas hydrate systems such as Hydrate Ridge or Walker Ridge and approximating these complex 2-D systems as locally 1-D system. This allows simple inputs for estimating hydrate saturation without requiring complex, transient, 2-D models. Thus, our model can be used to test several scenarios of gas hydrate occurrence and validate actual field observations, specifically (a) massive hydrates found in faults and fracture systems but not in surrounding sediments as in Hydrate Ridge [e.g., Tréhu et al., 2003, 2004a], (b) hydrate accumulation in high-permeability interbedded sand layers but not in the neighboring low-permeability layers as in Walker Ridge [Boswell et al., 2012a, 2012b], and (c) lack of gas hydrates in high-permeability horizontal layers despite significant methane flux. Another scenario where our model could be applied is when free gas columns are trapped in coarse-grain sequences beneath the GHSZ until overpressure forces the connected gas columns to migrate through the GHSZ and lead to coexistence of massive gas hydrates and free gas in saline pore waters [Tréhu et al., 2004b; Milkov et al., 2004]. For example, the average free gas saturations in Hydrate Ridge have been estimated to be 40–80% in the fractured sediment, significantly higher than values in the surrounding shale formation [Tréhu et al., 2004b; Daige et al., 2011]. Similarly, Boswell et al. [2012b] reported 50–90% hydrates concentrated within the coarse-grain, sand layers at Walker Ridge Block 313. With our 2-D model approach, we can explain such variability in hydrate distribution but also simultaneously determine the increased saturations within these high-permeability conduits and low saturations in the background matrix with the help of local transport parameters (flux, diffusivity, etc.) and the local permeability map of the reservoirs.

Three approximations used in our modeling are described above and reiterated here for clarity. First, we assume constant pore water salinity although gas hydrate formation may cause local elevation in salinity and impact the thermodynamic equilibrium. However, salinity changes can be assumed negligible in our quasi steady state model which further allows free gas to migrate into the GHSZ and convert to hydrate phase.
In order to model the coexistence of gas hydrate and free gas phases in the GHSZ, our current model needs to track pore water salt (Cl$^-$/C0) distribution over space and time and couple salinity effects due to hydrate formation and dissociation to thermodynamic phase calculations [Chatterjee, 2012]. Second, fracture formation in porous media is not modeled in our simulations. Gas hydrate formation results in permeability reduction and increased capillary entry pressure which exerts a barrier for pore fluids and free gas to flow. This leads to increased pore pressures and eventual sediment deformation and fracture genesis when the gas pressure equals total vertical stress. In our modeling, we utilize existing fracture networks and do not form or grow fractures as a result of gas hydrate accumulation. Third, gas hydrate accumulation around high-permeability layers or fracture systems is usually inhibited by competing forces of increased capillary entry pressures for free gas, gas hydrate formation in small pores, and lack of fluid migration [e.g., Anderson et al., 2003; Clennell et al., 1999; Henry et al., 1999]. At short timescales or at very small spatial scales these effects could lead to heterogeneity at smaller scales. However, such end-effects can be ignored in our modeling because we simulate near steady state processes over geologic lengths and timescales.

7. Conclusions

A 2-D model was developed to simulate gas hydrate and free gas accumulation in heterogeneous marine sediment. The model allows incorporation of lithologic heterogeneity, locally derived biogenic methane, deep-sourced external methane, and lateral fluid flow in the system. Key findings of this paper characterize the impact that high-permeability zones (e.g., vertical fractures and sand layers) have on gas hydrate and free gas distribution by focusing fluid flow along these fracture systems in the presence biogenic and/or thermogenic methane sources. These models generally quantify the effects of flow focusing.

Focused fluid flow through a vertical fracture network or dipping high-permeability sand layers increase local hydrate accumulation and saturation. These preliminary results, however, serve as a starting point and demonstrate that heterogeneity in natural gas hydrate systems is important for controlling gas hydrate and free gas saturations, and that these systems can be modeled. In this 2-D work, we show that the localized, focused, vertical, advective flux relative to diffusion ($Pe_{local}$) determines the magnitude of local hydrate and free gas saturation. Average local Peclet number is related to the average hydrate flux ($Pe_1 < S_n$) within high-permeability conduits, which compare favorably with the previous 1-D correlation.

Simulations with specified fluid flux and methane input from deeper sources, allow comparison of local methanogenesis and deeper methane sources on flow pathways and gas hydrate and/or free gas accumulation. Increased fluid flux from deeper external sources results in increased concentrations of hydrate and free gas. Permeability anisotropy, with a lower ratio of vertical to horizontal permeability show enhanced hydrate concentrations within the high-permeability conduits because anisotropy focuses more methane-charged fluid into these conduits. The 2-D model results quantify how focused fluid flow through high-permeability zones affects regional and local hydrate accumulation and saturation.

Dip angle of the permeable structure plays an important role in addition to lithology and stratigraphy because it influences vertical flux. We show that the vertical component of the fluid flow relative to the methane solubility gradient is the key driver of gas hydrate saturation and distribution in lithologically complex systems. Through a series of rigorous, 2-D, heterogeneous models, we show that flow focusing helps transport more methane through/to the GHSZ, and the vertical component of flow defines the gas saturation similar to our previous 1-D models. At a given location, regional and local gas hydrate saturation can be evaluated given the vertical flux and solubility gradient constraints.

Appendix A: Dimensionless Equations

A1. Initial Porosity Profile

At hydrostatic pressure, the porosity profile can be calculated as an analytical expression to serve as an initial condition:

$$\bar{\phi} = \frac{\eta}{\eta + (1 - \eta) \exp \left[ \frac{2\eta - 1}{\eta(1 + \eta)} \right]} \quad (A1)$$
A2. Water Mass Balance

\[
\left(1 + \gamma \frac{\tilde{\phi}}{\phi}\right) \frac{\partial}{\partial t} \left[ S_w \phi S_w + \epsilon \phi c_h \rho_h \right] + \frac{\tilde{\phi}}{N_{sc}} \left[ S_w \phi S_w + \epsilon \phi c_h \rho_h \right] \frac{\partial}{\partial t} \left[ \rho_w \frac{\partial}{\partial Z} \left( \rho_w \frac{\partial}{\partial Z} - \tilde{\sigma}_v \right) \right] + Pe \left(1 + \gamma \right) \frac{\partial}{\partial Z} \left[ \left(1 + \gamma \frac{\tilde{\phi}}{\phi}\right) \left( S_w \phi S_w + \epsilon \phi c_h \rho_h \right) \rho_w \frac{\partial}{\partial Z} \left( \rho_w \frac{\partial}{\partial Z} - \tilde{\sigma}_v \right) \right] = 0
\]

(A2)

The initial condition (I.C.) is assumed to be hydrostatic. The boundary condition (B.C.) at the seafloor is assumed hydrostatic, and the left and right boundaries of the domain are considered as no-flow boundaries. The initial condition and boundary conditions are expressed as follows:

I.C. : \[ \rho_w(\bar{Z}, \bar{x}, 0) = \frac{\rho_w g L_o + \rho_w g Z}{\rho_w g L_t} = \frac{L_o + Z}{L_t} = \frac{L_o + \bar{Z}}{L_t}, \quad \text{(Hydrostatic conditions)} \]  

(A3)

B.C.(1) : \[ \rho_w(0, \bar{x}, \bar{T}) = \frac{L_o}{L_t}, \quad \text{(Seafloor conditions)} \]  

(A4)

B.C.(2) : \[ \frac{\partial}{\partial Z} \left[ \rho_w \frac{\partial}{\partial Z} \left( \rho_w \frac{\partial}{\partial Z} - \tilde{\sigma}_v \right) \right] = 0 \quad \text{(No-flow conditions)} \]  

(A5)

The pore pressures in the 2-D model are not forced as hydrostatic, so the pressure gradient at the lowermost boundary cannot be modeled as a hydrostatic pressure gradient \[ \frac{\partial \tilde{\rho}_w}{\partial Z} (L_z, \bar{x}, \bar{T}) = 1. \] The bottommost boundary has been modeled by specifying the fluid flux and thereby specifying a non-hydrostatic pressure gradient. The specified fluid flux is equal to \[ U_{\text{sed}} + U_{\text{ext}} \] (for biogenic sources only, \( U_{\text{ext}} = 0 \)). Rewriting Darcy water flux (equation (5)) for the lowermost boundary as follows:

\[
v_w S_w \phi = v_s S_w \phi - \frac{k_{kw}}{\mu_w} \cdot (\nabla \rho_w - \rho_w g \nabla Z) = U_{\text{sed}} + U_{\text{ext}}
\]

(A6)

On normalizing,

\[
\tilde{v}_w S_w \frac{1 + \gamma \frac{\tilde{\phi}}{\phi}}{\phi} = \tilde{v}_s S_w \frac{1 + \gamma \frac{\tilde{\phi}}{\phi}}{\phi} - k_{kw} \left(1 + \frac{\gamma}{\phi}\right) \left( \frac{k_{kw} \rho_w g}{S_w \mu_w} \right) \cdot (\nabla \rho_w - \nabla Z) = \frac{1 + \gamma}{\phi} \left[ U_{\text{sed}} + U_{\text{ext}} \right]
\]

(A7)

Substituting equations (23) and (25), normalizing the term on the right and rearranging

\[
-N_{sc} \left(1 + \frac{\gamma}{\phi}\right) \left( \frac{k}{k_{sc}} \right) k_{kw} \cdot (\nabla \rho_w - \nabla Z) = \frac{[Pe_1 + Pe_2]}{Pe_1 \gamma/(1 - \eta)} - \frac{1 + \gamma \frac{\tilde{\phi}}{\phi}}{k_{kw}} S_w \tilde{v}_s
\]

(A8)

The non-hydrostatic pressure gradient at the bottom boundary is expressed using equation (A8):

\[
\frac{\partial \tilde{\rho}_w}{\partial Z} (L_z, \bar{x}, \bar{T}) = 1 - \frac{[Pe_1 + Pe_2]}{Pe_1 \gamma/(1 - \eta)} - \frac{1 + \gamma \frac{\tilde{\phi}}{\phi}}{k_{kw}} S_w \tilde{v}_s \quad \text{(Non-hydrostatic pressure gradient)}
\]

(A9)

where \( L_o \) is the seafloor depth, \( L_s \) is the width of the domain, and \( L_z \) is the depth to the bottom of the domain. Notably, the relative permeability of water at this boundary is unity, \( k_{kw}(L_z) = 1 \). Thus, a non-hydrostatic boundary condition at the lowermost boundary (equation (A9)) is derived by specifying a finite fluid flux at the boundary. Infinite \( N_{sc} \) represents infinite permeability, which implies hydrostatic conditions. For \( N_{sc} = \infty \), equation (A9) reduces the pressure gradient to unity which corresponds to a hydrostatic pressure gradient at the lowermost boundary of the simulation domain.

A3. Sediment Mass Balance

\[
- \frac{\tilde{\phi}}{N_{sc}} \frac{\partial}{\partial t} \left( \rho_w \frac{\partial}{\partial Z} - \tilde{\sigma}_v \right) + Pe_1 \left(1 + \frac{\gamma}{\phi}\right) \frac{\partial}{\partial Z} \left[ \left(1 - \frac{\tilde{\phi}}{\phi}\right) \tilde{\sigma}_v \right] = 0
\]

(A10)
The initial sediment velocity profile is evaluated assuming hydrostatic pressure, whereas the boundary condition for velocity of sediment at the seafloor is the normalized sedimentation rate and equal to unity.

\[
\text{I.C.} : \quad \bar{v}_i(Z, \bar{x}, 0) = \left( \frac{1 - \eta}{1 - \phi} \right) \quad \text{(Hydrostatic condition)} \tag{A11}
\]

\[
\text{B.C.} : \quad \bar{v}_i(0, \bar{x}, \bar{T}) = 1 \quad \text{(Seafloor condition)} \tag{A12}
\]

**A4. Organic Mass Balance**

\[
-\frac{\partial \bar{\rho}}{N_{t0} \sigma_t} \frac{\partial}{\partial t} \left( \bar{P}_w - \bar{\sigma}_v \right) + \left( 1 - \phi \right) \left( \frac{\partial \bar{\rho}}{\partial t} \right) \quad \text{Pe} \left( \frac{1 + y}{1 - \eta} \right) \left( \frac{\partial \bar{\rho}}{\partial t} \right) (1 - \phi) \bar{v}_i \bar{\alpha} = -D \alpha \left( 1 - \phi \right) \bar{\alpha} \tag{A13}
\]

Initially, there is no organic carbon present within the sediment, whereas the boundary condition of organic concentration at the seafloor is normalized to unity.

\[
\text{I.C.} : \quad \bar{\alpha}(Z, \bar{x}, 0) = 0 \tag{A14}
\]

\[
\text{B.C.} : \quad \bar{\alpha}(0, \bar{x}, \bar{T}) = 1 \quad \text{(Seafloor conditions)} \tag{A15}
\]

**A5. Methane Mass Balance**

\[
\left( \frac{1 + \gamma}{\gamma} \right) \frac{\partial}{\partial t} \left[ S_w \bar{v}_{l,m} + S_h \bar{v}_{l,m} \bar{\rho}_h + S_g \bar{v}_{l,m} \bar{\rho}_g \right] + \frac{\partial}{\partial x} \left[ S_w \bar{v}_{m} + S_h \bar{v}_{m} \bar{\rho}_h + S_g \bar{v}_{m} \bar{\rho}_g \right] \frac{\partial \bar{\rho}_w}{\partial t} - \frac{\partial \bar{\rho}_x}{\partial t} \\
\text{Pe} \left( \frac{1 + y}{1 - \eta} \right) \frac{\partial}{\partial t} \left[ \frac{1 + \gamma}{\gamma} \left( S_w \bar{v}_{l,m} + S_h \bar{v}_{l,m} \bar{\rho}_h + S_g \bar{v}_{l,m} \bar{\rho}_g \right) \right] - N_{w} k_{rw} \left( \frac{1 + \gamma}{\gamma} \right) \left( \frac{1 + \gamma}{\gamma} \right) \left( \frac{\partial \bar{\rho}_w}{\partial z} - 1 \right) \bar{v}_m \\
\text{Pe} \left( \frac{1 + y}{1 - \eta} \right) \frac{\partial}{\partial t} \left[ \frac{1 + \gamma}{\gamma} \left( S_w \bar{v}_{m} + S_h \bar{v}_{m} \bar{\rho}_h + S_g \bar{v}_{m} \bar{\rho}_g \right) \right] - N_{w} k_{rw} \left( \frac{1 + \gamma}{\gamma} \right) \left( \frac{1 + \gamma}{\gamma} \right) \left( \frac{\partial \bar{\rho}_w}{\partial z} \right) \bar{\rho}_g \bar{v}_m \\
= \frac{\partial}{\partial z} \left[ \left( \frac{1 + \gamma}{\gamma} \right) S_w \bar{v}_{l,m} \frac{\partial \bar{\rho}_w}{\partial z} + \frac{\partial}{\partial x} \left[ \left( \frac{1 + \gamma}{\gamma} \right) S_w \bar{v}_{m} \frac{\partial \bar{\rho}_w}{\partial x} \right] + M_{\text{org}} \rho \bar{\rho}_{\text{ext}} \right] - D \alpha \left( 1 - \phi \right) \bar{\beta} \tag{A16}
\]

Initially, there is no methane in the system. At the seafloor, methane concentration is equal to zero; methane flux is set to zero at the right and left boundaries of the domain; the bottom boundary has a choice of boundary condition, depending on the methane source. The methane concentration gradient is equal to zero for a system with biogenic sources only (equation (A20)), whereas the methane concentration is set to be a constant value for deeper methane sources (equation (A21)).

\[
\text{I.C.} : \quad \bar{\psi}_{l,m}(Z, \bar{x}, 0) = 0 \tag{A17}
\]

\[
\text{B.C.}(1) : \quad \bar{\psi}_{l,m}(0, \bar{x}, \bar{T}) = 0 \quad \text{(Seafloor condition)} \tag{A18}
\]

\[
\text{B.C.}(2) : \quad \frac{\partial \bar{\psi}_{l,m}}{\partial x}(0, \bar{x}, \bar{T}) = \frac{\partial \bar{\psi}_{l,m}}{\partial x}(Z, L_x, \bar{T}) = 0 \quad \text{(No – flow conditions)} \tag{A19}
\]

\[
\text{B.C.}(3) : \quad \frac{\partial \bar{\psi}_{l,m}}{\partial x}(L_z, \bar{x}, \bar{T}) = 0 \quad \text{(Biogenic in situ sources only)} \tag{A20}
\]

\[
\text{B.C.}(4) : \quad \bar{\psi}_{l,m}(L_z, \bar{x}, \bar{T}) = \bar{\psi}_{l,m,\text{ext}} \quad \text{(Thermogenic sources only)} \tag{A21}
\]
Appendix B: Time Derivative Term Expansion

The accumulation terms in the mass balance equations are basically a product of primary variables like saturation ($S_j$), porosity ($\phi$), and mass fraction ($c_i$) of component $i$ in phase $j$. The time derivative can be expanded as shown:

$$\Delta_t \left( \phi S_j c_i \right) = \phi^{n+1} S_j^{n+1} c_i^{n+1} - \phi^n S_j^n c_i^n$$ (B1)

where $\Delta_t$ represents the change in time and superscripts $n+1$ and $n$ represent different time steps. On adding and subtracting a term $\phi^{n+1} S_j^n c_i^n$:

$$\Delta_t \left( \phi S_j c_i \right) = \phi^{n+1} S_j^{n+1} c_i^{n+1} - \phi^n S_j^n c_i^n + \phi^{n+1} S_j^n c_i^n - \phi^n S_j^n c_i^n$$ (B2)

On rearranging:

$$\Delta_t \left( \phi S_j c_i \right) = \phi^{n+1} \left( S_j^{n+1} c_i^{n+1} - S_j^n c_i^n \right) + S_j^n c_i^n \left( \phi^{n+1} - \phi^n \right)$$ (B3)

which is restated as:

$$\Delta_t \left( \phi S_j c_i \right) = \phi^{n+1} \Delta_t \left( S_j c_i \right) + S_j^n c_i^n \left( \phi^{n+1} - \phi^n \right)$$ (B4)

Porosity is a function of effective stress (equation (18)) and is normalized using the scaling scheme (equations (20–33)). Reduced porosity $\tilde{\phi}$ is related to the dimensionless lithostatic stress $\tilde{\sigma}_v$ and dimensionless pore pressure $\tilde{p}_w$:

$$\tilde{\phi} = -\eta \exp \left[ \frac{\tilde{\sigma}_v - \tilde{p}_w}{N_1 \phi} \right]$$ (B5)

Time derivative of porosity as a function of pore pressure can be written as follows:

$$\Delta_t \left( \phi S_j c_i \right) = \phi^{n+1} \Delta_t \left( S_j c_i \right) + S_j^n c_i^n \frac{\partial \phi}{\partial \tilde{p}_w} \Delta_t \tilde{p}_w$$ (B6)

From the effective stress-porosity relationship, porosity is differentiated with respect to pore pressure.

$$\Delta_t \left( \phi S_j c_i \right) = \phi^{n+1} \Delta_t \left( S_j c_i \right) + S_j^n c_i^n \left[ \frac{\phi - \phi_w}{\sigma_\phi} \right] \left[ 1 - \frac{\sigma^{n+1}_j - \sigma^n_j}{\tilde{p}_w^{n+1} - \tilde{p}_w^n} \right] \Delta_t \tilde{p}_w$$ (B7)

The time derivative expansion for accumulation terms in the following mass balance equations can be expanded using the same technique. These time derivative terms are now substituted back in the mass balance equations (equations (1–4)) and then normalized with the help of the scaled variables discussed above (equations (20–33)).

Appendix C: Analytical Theory Relating the Fluid Flux and Average Hydrate Saturation

An analytical theory is developed to relate the average hydrate saturation to the fluid flux through porous media in any orientation to the horizontal axis (Figure C1). This theory revisits the approach developed by Bhatnagar et al. [2011] to correlate the depth of the Sulfate methane transition (SMT) to the average hydrate saturation in marine sediments dominated by methane rising with pore fluids from depth. Likewise, in this model, it is assumed that there is no microbial generation of methane, and
methane-charged pore fluids rising from depth is the only source of methane. Furthermore, pore water sulfate is assumed to be absent in pore fluids, and methane can escape into the seafloor even though some of the detailed pore water chemistry models have been published in the last decade relating sulfate and methane fluxes at the SMT in shallow sediments [e.g., Davie and Buffett, 2003b; Bhatnagar et al., 2008a, 2011; Chatterjee et al., 2011a; Malinverno and Pohlman, 2011].

**C1. Water Mass Balance**

For simplicity, steady state water mass balance is expressed as

\[ \frac{d}{dz} \left[ U_{i,\text{vert}} C_{w} \rho_{w} + U_{i} \left( 1 - \phi \right) \phi S_{h} C_{h} \rho_{h} \right] = 0, \quad 0 < z < L_{t} \]  

(C1)

The invariant steady state water flux can be restated as

\[ U_{i,\text{vert}} C_{w} \rho_{w} + U_{i} \left( 1 - \phi \right) \phi S_{h} C_{h} \rho_{h} = F_{w,0} = (U_{\text{sed}} + U_{\text{ext}} \sin \theta) \rho_{w}, \quad 0 < z < L_{t} \]  

(C2)

where \( \theta \) is the dip angle relative to the horizontal axis. The net vertical fluid (liquid) flux, \( U_{i,\text{vert}} \), sediment flux, \( U_{\text{sed}} \), and the fluid flux due to sedimentation and compaction, \( U_{\text{sed}} \), are assumed to be vertical; whereas the external fluid flow, \( U_{\text{ext}} \), is along the permeable conduit inclined at angle \( \theta \) to the horizontal. Therefore, the vertical component of the external fluid flux, \( U_{\text{ext}} \sin \theta \), is added to the sedimentation-compaction-driven fluid flux, \( U_{\text{sed}} \), to obtain the net vertical fluid flux. Assuming low methane solubility in water, \( C_{w} \) is approximated as unity and normalizing gas hydrate density by water density:

\[ U_{i,\text{vert}} = (U_{\text{sed}} + U_{\text{ext}} \sin \theta) - \frac{U_{i}}{1 - \phi} \phi S_{h} C_{h} \rho_{h}, \quad 0 < z < 1 \]  

(C3)

**C2. Methane Mass Balance**

The steady state, two-phase (dissolved methane and hydrate) methane mass balance between the seafloor \((z = 0)\) and the base of the GHSZ \((z = L_{t})\) is expressed assuming \( S_{g} = 0 \) and no methane formation by methanogenesis reaction:

\[ \frac{d}{dz} \left[ U_{i,\text{vert}} C_{w} \rho_{w} + U_{i} \left( 1 - \phi \right) \phi S_{h} C_{h} \rho_{h} - \phi (1 - S_{h}) \rho_{w} D_{m} \frac{d C_{m}^{l}}{dz} \right] \sin \theta = 0, \quad 0 < z < L_{t} \]  

(C4)

where \( U_{i,\text{vert}} \) is the total vertical water flux, \( U_{i} \) is the sediment flux, \( S_{h} \) is the gas hydrate saturation, and \( C_{m}^{l} \) is the mass fraction of methane in the hydrate phase (a constant, \( C_{m}^{l} = 0.134 \)) for structure I hydrate [Sloan and Koh, 2007]. Therefore, steady state methane mass flux can be rewritten as

\[ U_{i,\text{vert}} C_{w} \rho_{w} + U_{i} \left( 1 - \phi \right) \phi S_{h} C_{h} \rho_{h} - \phi (1 - S_{h}) \rho_{w} D_{m} \frac{d C_{m}^{l}}{dz} \sin \theta = F_{\text{Ch}_{4}} \rho_{w}, \quad 0 < z < L_{t} \]  

(C5)

where \( F_{\text{Ch}_{4}} \) is the methane mass flux. Using the same scaling scheme discussed above and in Bhatnagar et al. [2007, 2011], sediment flux is scaled by \( U_{\text{sed}} \), methane mass fractions by methane solubility at the base of the GHSZ \((C_{m,\text{eqb}})\) and substituting \( U_{i,\text{vert}} \) as in equation (C3). The scaled methane mass flux is

\[ Q C_{m}^{l} + \frac{P_{e_{1}} U_{i}}{1 - \phi} \left( \frac{1 + \gamma}{\gamma} \right) S_{h} \rho_{h} \left( C_{m}^{l} - C_{m}^{l, \text{eqb}} \right) \]

\[ - \left( 1 + \gamma \phi \right) (1 - S_{h}) \frac{d C_{m}^{l}}{dz} \sin \theta = F_{\text{Ch}_{4}} L_{t} D_{m} C_{m,\text{eqb}} \left( \frac{1 + \gamma}{\gamma} \right) = f_{\text{Ch}_{4}} \rho_{w}, \quad 0 < z < 1 \]  

(C6)

where \( f_{\text{Ch}_{4}} \) is normalized methane mass flux and \( Q \) is defined as

\[ Q = \left( \frac{1 + \gamma}{\gamma} \right) (P_{e_{1}} + P_{e_{2}} \sin \theta) \]  

(C7)
The depth domain can be divided into two distinct regions: between the seafloor and the shallowest occurrence of gas hydrate, \(1 - \bar{L}_h\) and from this depth to the base of the GHSZ where \(\bar{L}_h\) is the thickness of the gas hydrate occurrence zone (GHOZ). Normalized methane mass flux (equation (C6)) is rewritten for these two zones with simplified expressions as discussed in Bhatnagar et al. [2011]. For the lower region \(1 - \bar{L}_h < \bar{Z} < 1\), hydrate saturation, \(S_h\) is set to zero as \(\bar{Z} \to 1 - \bar{L}_h\):

\[
f_{\text{CH}_4} = \frac{Q \bar{c}_{m,\text{sol}} (1 - \bar{L}_h)}{1 - \exp \left( \frac{Q}{\sin \theta} [g(1 - \bar{L}_h) - g(0)] \right)} , \quad 0 < \bar{Z} < 1 - \bar{L}_h \tag{C8}
\]

\[
f_{\text{CH}_4} = Q \bar{c}_{m,\text{sol}}(\bar{Z}) - \left( \frac{1 + \gamma \phi}{\gamma} \right) \bar{c}_{m,\text{sol}}(\bar{Z}) \sin \theta , \quad 1 - \bar{L}_h < \bar{Z} < 1
\]

where \(\bar{c}_{m,\text{sol}}(\bar{Z})\) is the methane solubility at any scaled depth, \(\bar{Z}\) and is defined as a single-parameter model, \(\bar{c}_{m,\text{sol}}(\bar{Z}) = \exp(-r_z(1 - \bar{Z}))\) as shown in Bhatnagar et al. [2011] and \(g(\bar{Z})\) represents the integral of the porosity term and is defined as

\[
g(\bar{Z}) = \int_0^{1 - \bar{L}_h} \left( \frac{\gamma}{1 + \gamma \phi} \right) d\bar{Z} = \frac{N_{ch}}{1 + \gamma \phi} \bar{Z} + \eta^2 \ln \left[ \frac{\eta (1 + \gamma) + (1 - \eta) e^{2N_{ch}}}{(1 + \gamma)N_{ch}} \right] \tag{C9}
\]

The unknown parameter, \(\bar{L}_h\) is solved by equating the two methane mass fluxes (equation (C8)) in the two distinct zones at \(\bar{Z} = 1 - \bar{L}_h\) for different values of \(Q\).

Rearranging equation (C6) and following the theory discussed in Bhatnagar et al. [2011], the gas hydrate profile is rewritten as a function of the scaled depth, \(\bar{Z}\) as follows:

\[
S_h(\bar{Z}) = \frac{f_{\text{CH}_4} - \bar{c}_{m,\text{sol}}(\bar{Z})}{\bar{c}_{m,h} + \bar{c}_{m,\text{sol}}(\bar{Z}) \sin \theta} , \quad 1 - \bar{L}_h < \bar{Z} < 1 \tag{C10}
\]

The mass fraction part of the first term in the denominator of equation (C10), \((\bar{c}_{m,h} - \bar{c}_{m,\text{sol}}(\bar{Z}))\) can be simplified further. In the main text, \(\bar{c}_{m,h}\) was defined as \(\bar{c}_{m,h} = \bar{c}_{m,h} / \bar{c}_{m,\text{eqht}}\) which for most marine systems is of the order of \(\bar{c}_{m,h} = 0.134/10^{-3} = 10^2\). The remaining terms, \(\bar{c}_{m,\text{sol}}(\bar{Z})\) and \(\bar{c}_{m,\text{sol}}(\bar{Z})\), are usually less than unity implying that \(\bar{c}_{m,h}\) is approximately 2 orders of magnitude greater than the remaining terms in the denominator. Thus, this approximates and simplifies equation (C10) as

\[
S_h(\bar{Z}) \approx \frac{f_{\text{CH}_4} - \bar{c}_{m,\text{sol}}(\bar{Z})}{\bar{c}_{m,h} + \bar{c}_{m,\text{sol}}(\bar{Z}) \sin \theta} , \quad 1 - \bar{L}_h < \bar{Z} < 1 \tag{C11}
\]

Integrating this saturation profile over the entire depth between the seafloor and the base of GHSZ and multiplying the integral by \(P_{e_1}\), average hydrate flux, \(P_{e_1}(S_h)\) can be defined as a function of system parameters:

\[
P_{e_1}(S_h) \approx \int_{1 - \bar{L}_h}^{1} \left( \frac{f_{\text{CH}_4} - \bar{c}_{m,\text{sol}}(\bar{Z})}{1 - \bar{L}_h} \right) \frac{1}{\bar{c}_{m,h} + \bar{c}_{m,\text{sol}}(\bar{Z}) \sin \theta} d\bar{Z} \tag{C12}
\]

Specifying the dip angle of the configuration (\(\theta\)), and other system parameters, and varying external fluid flux, \(P_{e_2}\), top of gas hydrate zone, \(1 - \bar{L}_h\) is first computed. Using this value and other system parameters, average hydrate flux \(P_{e_1}(S_h)\) is evaluated. The average hydrate saturation, \(\langle S_h \rangle\), can be easily computed from this product since \(P_{e_1}\) is a constant parameter at a given geologic setting as it depends on sedimentation rate at the seafloor. Therefore, the fluid flux within a flow conduit in any orientation (to the horizontal axis) is related to the average hydrate saturation in these gas hydrate systems using a simple analytical formulation.
Notations

- $c_i$: Mass fraction of component $i$ in phase $j$
- $c_{i,m,eqb}$: CH$_4$ solubility at the base of the GHSZ
- $Da$: Damköhler number
- $D_m$: Diffusivity of CH$_4$ in seawater
- $g$: Acceleration due to gravity
- $k$: Absolute sediment permeability tensor
- $k_{o,j}$: Absolute sediment permeability at seafloor
- $k_{r,j}$: Relative permeability of phase $j$
- $k_{r,j}^{e}$: End-point value of $k_{r,j}$
- $L_o$: Depth of the seafloor ($\bar{z} = 0$)
- $L_1$: Depth to the base of the GHSZ
- $L_2$: Depth to the base of the domain
- $L_x$: Width of the domain
- $L_{\phi}$: Characteristic depth of compaction
- $M_i$: Molecular weight of component $i$
- $N_{sc}$: Sedimentation-compaction group
- $N_{t,\phi}$: Porosity and compaction group
- $n$: Pore size distribution index
- $Pe_1$: First Peclet number
- $Pe_2$: Second Peclet number
- $P_i$: Pressure of phase $j$
- $P_c$: Capillary pressure
- $P_{c,0}$: Capillary pressure at reference value
- $P_{c,e,0}$: Capillary entry pressure at reference value
- $S_j$: Saturation of phase $j$
- $S_{r,j}$: Residual saturation of phase $j$
- $S$: Sedimentation rate
- $t$: Time
- $U_{l,sed}$: Fluid flux due to sedimentation
- $U_{l,ext}$: Fluid flux due to external fluid flow
- $V_j$: Velocity of phase $j$
- $z$: Depth below the seafloor
- $\alpha$: Organic carbon content in sediment
- $\alpha_o$: Organic carbon content at the seafloor
- $\beta$: Normalized carbon content at the seafloor
- $\eta$, $\gamma$: Reduced porosity parameters
- $\lambda$: Kinetic rate constant for methanogenesis
- $\mu_j$: Viscosity of phase $j$
- $\rho_j$: Density of phase $j$
- $\sigma_{\phi}$: Total vertical stress
- $\sigma_{d,\phi}$: Characteristic stress of compaction
- $\sigma_{gw}$: Interfacial tension at gas-water contact
- $\theta$: Gas-water contact angle
- $\theta$: Dip angle
- $\phi$: Porosity
- $\phi_o$: Porosity at the seafloor
- $\phi_{\infty}$: Porosity at infinite depth

Subscripts/Superscripts

- $g$, $l$, $h$, $s$: Gas, liquid, hydrate, and sediment phases
- $m$, $w$: CH$_4$ and water components
References


