# Large-Scale Simulation of Methane Hydrate Dissociation along the West

# Spitsbergen Margin

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### Abstract

Vast quantities of methane are trapped in oceanic hydrate deposits, and there is concern that a rise in the ocean temperature will induce dissociation of these hydrate accumulations, potentially releasing large amounts of methane into the atmosphere. The recent discovery of active methane gas venting along the landward limit of the gas hydrate stability zone (GHSZ) on the shallow continental slope west of Spitsbergen could be an indication of this process, if the source of the methane can be confidently attributed to dissociating hydrates. In the first largescale simulation study of its kind, we simulate shallow hydrate dissociation in conditions representative of the West Spitsbergen margin to test the hypothesis that the observed gas release originated from hydrates. The simulation results are consistent with this hypothesis, and are in remarkable agreement with the recently published observations. They show that shallow, lowsaturation hydrate deposits, when subjected to temperature increases at the seafloor, can release significant quantities of methane, and that the releases will be localized near the landward limit of the top of the GHSZ. These results indicate the possibility that hydrate dissociation and methane release may be both a consequence and a cause of climate change.

Index terms: 3004, 1600, 1800, 1605, 4902

Keywords: gas hydrates, methane clathrates, abrupt change

Gas hydrates are solid crystalline compounds in which gas molecules are lodged within a clathrate crystal lattice (Sloan and Koh, 2007). Vast quantities of methane are trapped in oceanic hydrate deposits. Initial investigations estimated the total amount of methane hydrate currently residing in the oceans at 10,000 gigatons (Gt) of methane carbon (Gornitz and Fung, 1994; Kvenvolden, 1999; Borowski, 2004), but more recent studies have produced widely different methane hydrate estimates—ranging from an upper estimate of 27,300 Gt in hydrates along continental margins (74,400 Gt globally) (Klauda and Sandler, 2005) to a lower estimate of 3,000 Gt in hydrates (Buffett and Archer, 2004). The depth at which hydrates remain stable depends on the pressure (as imposed by water depth) and temperature. An increase in water temperature at the seafloor would change the extent of the gas hydrate stability zone (GHSZ) and possibly induce hydrate dissociation, leading to release of methane. Recent computational studies quantified the potential for local methane release under warming conditions (Reagan and Moridis, 2008) using a 1-D representation of sub-seafloor processes. Because methane is a powerful greenhouse gas, there is considerable concern that a rise in the ocean temperature can induce oceanic hydrate dissociation, with adverse climatic consequences. This positive-feedback mechanism has been proposed as a significant contributor to rapid climate changes in the late Quaternary period (Kennett et al., 2000). While this hypothesis is controversial, and the relationship between hydrates and climate has not yet been established, the role of methane in climate cycles is currently an active area of research and hydrates are considered a potential source (Masciarelli, 2009).

Westbrook and collaborators recently discovered over 250 plumes of methane gas bubbles erupting from the seabed off the west Spitsbergen continental margin along the present landward limit of the GHSZ (Westbrook et al., 2009). Two possible explanations are proffered: 1) that the methane originates from dissociating shallow hydrates, and/or 2) that methane arriving from deeper sources has formed hydrate in the shallow GHSZ that blocks or channels (by reducing the sediment permeability) the ascending gaseous methane.

We conducted a large-scale, 2-D simulation of a shallow hydrate system in conditions representative of the Arctic continental shelf at the western Svalbard margin to test these hypotheses and assess the potential for hydrate dissociation, methane release, and plume formation as a result of ocean warming.

We simulate the system using the massively parallel version of the TOUGH+HYDRATE code (pT+H) (Moridis, 2003; Moridis et al., 2008), which models the fully coupled phase behavior, flow of fluids and heat, and nonisothermal hydration reaction in complex geologic media across multiple spatial dimensions. The serial version of TOUGH+HYDRATE has been used in earlier, 1-D investigations of hydrate dissociation in response to ocean temperature change (Reagan and Moridis, 2008) as well as numerous 1-D and 2-D gas hydrate reservoir engineering studies (Moridis et al., 2009; Moridis and Sloan, 2007). This study is the first application of pT+H to a climate-driven system of this magnitude.

The model system is a 2-D slice of a representative slope (Figure 1) 5000 m in length. The western Svalbard continental shelf has a 3%–5% slope, as indicated by local averages from GEBCO bathymetry data for the western Svalbard region (GEBCO, 1997) and as reported by Westbrook (2009) in the region of methane plume formation. We select a 5% grade, encompassing the seafloor in the region of methane plume formation, with the top of the slope at 300 m depth and extending downslope to 550 m depth. The region of sediments modeled extends 300 m below the seafloor. Figure 1 also shows a schematic of the mesh and the initial extent of the GHSZ. To represent the system at a suitable level of detail, we use 300,000 gridblocks (1000×300, resulting in 1,200,000 coupled equations for the hydrate-water-inhibitor system), with a horizontal discretization of dx = 5 m, a 2D slice thickness of dy = 1 m, and a variable vertical discretization, beginning with dz=0.25 m from the seafloor to z =-50 m, dz = 0.5 m between z = -50 m and z = -75 m, and a logarithmic progression (dz = 0.5-15.8 m) from z = -75 to z = -300 m.

The initial conditions involve a geothermal gradient of 8.7 °C/100 m as taken from measurements further downslope (Haake et al., 2008), 3.5 wt% initial salinity, and a uniform initial hydrate saturation of 3% between z = -5 m and the base of the GHSZ. Heterogeneity of the hydrate layer is not considered in this study for lack of coring data or profiles. Sediment porosity,  $\phi = 0.55$ , reflects deeper locations further offshore (Haake et al., 2008), and an intrinsic sediment permeability of  $k = 10^{-15} \text{ m}^2$  (1 mD) is representative of unconsolidated ocean sediments and similar to previously simulated arctic scenarios (Reagan and Moridis, 2008). Other physical properties of the sediments and porous-media system are summarized elsewhere (Reagan and Moridis, 2008); however, this work uses a full 2-D representation of the model system. The extent of the GHSZ is computed directly from the depth and initial temperature distribution, and the initial state of the system was attained using pT+H, bringing the system to thermal, chemical, thermodynamic, and hydrostatic equilibrium at the initial temperature. A preexisting region of free gas, inferred (but not confirmed) to exist underneath such hydrate deposits, is not included in this simulation, as the actual quantity of gas is unknown, and we seek to quantify the  $CH_4$  that can be released due to hydrate dissociation alone. The top of the domain is an open boundary representing heat and mass transfer between the sediment and the overlying ocean. The pressure at the upper boundary is variable with depth but time-invariable, representing constant ocean levels (Reagan and Moridis, 2008). The bottom of the domain is a closed boundary beyond the expected range of temperature propagation on the timescales of this study (Reagan and Moridis, 2008) and is held at constant temperature corresponding to the initial geothermal gradient.

Recent climate simulations coupling ocean circulation, atmospheric circulation, and atmospheric chemistry (Meehl et al., 2007) indicate that, under current climate conditions and a 1%/yr increase in atmospheric CO2, the temperature at the seafloor would rise by 1°C over the

next 100 yr, and possibly by another 3°C in the following century. Previous work on the response of shallow hydrates to ocean temperature change (Reagan and Moridis, 2008) indicates that temperature changes as small as 1–3°C can have significant effects on shallow hydrates. For the western Spitsbergen continental shelf, historical temperature data suggest that a 3 °C change in bottom-water temperature has occurred over the last 100 years with an average rate of 0.03 °C /yr (Westbrook et al., 2009). We model this rapid change with a linear 3 °C temperature increase at the seafloor (at all depths from 300 to 550 m) over a 100 yr period (i.e., beginning roughly at 1908 CE) with an initial temperature of  $T_0 = 0$  °C at t = 0. After 100 years of simulation (corresponding to 2008 CE), the ocean temperature is held constant for an additional 100 yr (a conservative, best-case scenario, assuming no further changes in ocean temperature). Thus, in this study the total temperature change and total simulation time are limited, as extrapolation of recent temperature trends over centuries is difficult at best, and we are most interested in capturing century-scale phenomena that may already be occurring, and that may already be observable.

Figure 2 illustrates the evolution of hydrate saturation,  $S_H$ , with time. Over 200 yr, the upper (leftmost) extent of methane gas hydrate recedes approximately 1,500 m downslope. The effects of the lowering of the top of the GHSZ are evident, as a distinct upper dissociation front is visible at t = 50 yr, 100 yr, and 130 yr. Simulations of 1-D hydrate-bearing sediment columns under arctic conditions (Reagan and Moridis, 2008) have demonstrated that the rate of dissociation is regulated by heat transfer limitations, as hydrate dissociation is strongly endothermic, and thus we see the formation of a sharp dissociation front in both the previous 1-D study and in this 2-D simulation. Secondary hydrate is seen to form at the bottom of the hydrate-bearing sediments, locally increasing  $S_H$  beyond the initial value of 3% to 3.5% by t = 200 yr.

Figure 3 shows the evolution of gas saturation,  $S_G$ . At t = 50 yr, a large zone of free gas envelopes a localized plume of high gas saturation. The region of highest saturation coincides

with the bottom of the GHSZ, as dissociation-originating CH<sub>4</sub> ascends along the base of the hydrate. Note that this is adjacent to the region of secondary hydrate formation (increased  $S_H$ ) seen in Figure 2. This plume contacts the seafloor between x = 750 and x = 1500 m (340 m to 375 m depth), moving downslope with time.

Simulation outputs indicate that the first appearance of gaseous methane at the seafloor occurs around t = 80 yr (~1990 CE), and that the onset of gas release is rapid. In Figure 4, presents the evolution of gas flux,  $Q_{CH4}$  (mol/yr-m<sup>2</sup>) as a function of time and downslope position. The peaks of gaseous flux in Figure 4 correspond closely to the intersection between the seafloor boundary and the gas-phase plume pictured in Figure 3 at t = 100 yr, t = 130 yr, and t = 200 yr. The peaks also fall within the range of depths for plume origination reported by Westbrook *et al.* (2009).

The evolution of the total methane flux, in both aqueous and gaseous form, for the entire top interface of the 1 m-wide, 5000 m-long, 2-D slice of the system was recorded during the simulation. Methane release into the ocean begins at approximately t = 30 yr (in the dissolved phase) and reaches a peak of 8800 mol/yr at t = 175 yr, with a total of  $9.2 \times 10^5$  mol of CH<sub>4</sub> released by t = 200 yr. This corresponds to 141 kg/yr CH<sub>4</sub> per 1 m width of slope released solely due to dissociating hydrate. Integrated over the 30-km length of the plume-zone at the Spitsbergen margin, this would contribute 0.004 Tg/yr of CH<sub>4</sub> to the ocean from this hydrate system alone, small in comparison to the global atmospheric flux of methane but potentially significant to the chemistry and biology of the ocean in regions affected by such releases. The areal extent of Arctic Ocean shelf between 300 m and 500 m deep encompasses roughly 700,000 km<sup>2</sup> (Carmack and Wasserman, 2006), and this system represents a mere 0.02% of that area. If sparse-hydrate plume systems such as this exist in other areas with similar depth and temperature conditions (along the Arctic continental shelf, Bering Sea, or Okhotsk Sea), the cumulative effect could be proportionally larger and huge in absolute terms.

The simulations presented in this paper closely describe the type and extent of hydratebearing system thought to exist around the Svalbard margin, and allow us to make several conclusions about the nature of this system:

(1) The simulated release of  $CH_4$  by dissociating hydrates is consistent with observations the hypothesis that dissociating hydrate alone can provide a significant quantity of methane gas in ocean-temperature-driven methane release scenarios.

(2) The increase in  $S_H$  along the base of the GHSZ and the diversion of gas along the bottom of the hydrate deposit is consistent with the hypothesis that hydrate-bearing sediments can alter and direct the sub-seafloor transport of methane. This is due to the reduced permeability of hydrate-bearing sediments and the change in the relative permeability of sediments through the formation of new hydrate at the bottom of the GHSZ.

(3) Dissociation of shallow hydrates, combined with diversion of the hydrate-derived gas by hydrate-bearing sediments, may result in the formation of highly localized plumes of methane gas aligned with the landward limit of the receding GHSZ.

(4) The configuration of the simulated gas release at the ocean floor resembles the configuration of gas plumes observed off the Spitsbergen margin, with gas plumes originating at 340 m to 375 m depth.

### Notation

$\phi$	Porosity
$Q_{ m CH4}$	Methane flux (mol/yr-m <sup>2</sup> )
$S_H$	Hydrate saturation
$S_G$	Gas saturation
$T_0$	Initial temperature (°C)
t	time (yr)
x	downslope distance (m)

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#### References

- Borowski, W.S. (2004), A review of methane and gas hydrates in the dynamic, stratified system of the Blake Ridge region, offshore southeastern North *America. Chem. Geology*, 205, 311-346.
- Buffett, B., and D. Archer (2004), Global inventory of methane clathrate: Sensitivity to changes in environmental conditions, *Earth Planetary Sci. Lett.*, 227, 185-199.
- Carmack, E., and P. Wasserman (2006), Food webs and physical-biological coupling on pan-Arctic shelves: Unifying concepts and comprehensive perspectives, *Progress in Oceanography*, 71, 446-477.
- GEBCO (1997), Published by the British Oceanographic Data Centre on behalf of Intergovernmental Oceanographic Commission and International Hydrographic Organization.
- Gornitz V., and I. Fung (1994), Potential distribution of methane hydrate in the world's oceans, *Global Biogeochem. Cycles*, *8*, 335-347.
- Haake, R.R., Westbrook, G.K., and M.S. Riley (2008), Controls on the formation and stability of gas hydrate-related bottom-simulating reflectors (BSRs): A case study from the west Svalbard continental slope, *J. Geophys. Res.*, *113*, B05104, doi: 10.1029/2007JB005200.
- Jakobsson, M., Macnab, R., Mayer, L., Anderson, R., Edwards, M., Hatzky, J., Schenke, H. W., and P. Johnson (2008), An improved bathymetric portrayal of the Arctic Ocean: Implications

for ocean modeling and geological, geophysical and oceanographic analyses, *Geophys. Res. Lett.*, *35*, L07602, doi:10.1029/2008GL033520.

- Kennett, J.P., Cannariato, K.G., Hendy, L.L., and R.J. Behl (2000), Carbon isotopic evidence for methane hydrate instability during quaternary interstadials. *Science*, 288, 128-133.
- Klauda, J.B., and S.I. Sandler (2005), Global distribution of methane hydrate in ocean sediment. *Energy and Fuels*, *19*, 459-470.
- Kvenvolden, K.A. (1999), Potential effects of gas hydrate on human welfare, *Proc. Nat. Acad. Sci.*, *96*, 3420-3426.
- Mascarelli, A.L., A Sleeping Giant? (2009) *Nature Reports Climate Change*, *3*(4), doi: 10.1038/ climate.2009.24.
- Meehl, G., et al. (2007), *Global climate projections, Climate Change 2007: The Physical Basis*, 789-844, Cambridge Univ. Press, Cambridge, UK.
- Moridis, G.J. (2003), Numerical Studies of Gas Production from Methane Hydrates, *SPE Journal*, *32*(8).
- Moridis, G.J., and E.D. Sloan (2007), Gas production potential of disperse low-saturation hydrate accumulations in oceanic sediments, *Energy Conv. Manag.*, 48, 1834-1849, doi:10.1016/j.enconman.2007.01.023.
- Moridis, G.J., Kowalsky, M.B., and K. Pruess (2008), TOUGH+HYDRATE v1.0 User's Manual: A Code for the Simulation of System Behavior in Hydrate-Bearing Geologic Media, Report LBNL-0149E, Lawrence Berkeley National Laboratory, Berkeley, CA.
- Moridis, G.J., Reagan, M.T., Kim, S.J., Seol, Y., and K. Zhang (2009), Evaluation of the Gas Production Potential of Marine Hydrate Deposits in the Ulleung Basin of the Korean East Sea, SPE 110859, *SPE Journal*, doi:10.2118/110859-PA.
- Reagan, M. T., and G. J. Moridis (2008), Dynamic response of oceanic hydrate deposits to ocean temperature change, J. Geophys. Res., 113, C12023, doi:10.1029/2008JC004938.

- Sloan, E.D. and C. Koh (2007), *Clathrate Hydrates of Natural Gases*, 3<sup>rd</sup> Edition, CRC Press, New York, NY.
- Westbrook, G.K., Thatcher, K.E, Rohling, E.J., Piotrowski, A.M., Palike, H., Osborne, A.H.,
  Nisbet, E.G., Minshull, T.A., Lanoiselle, M., James, R.H., Huhnerbach, V., Green, D., Fisher,
  R.E., Crocker, A.J., Chabert, A., Bolton, C., Beszczynska-Möller, A., Berndt, C., and A.
  Aquilina (2009), Escape of methane gas from the seabed along the West Spitsbergen
  continental margin *Geophys. Res. Lett.*, *36*, L15608, doi: 10.1029/2009GL039191.

### **Figure Captions**

Figure 1. Illustration of the system location, simulated domain, extent of gas hydrate stability zone, and boundaries for the 2-D sloping system. Gridlines are a schematic representation only (not to scale). Map courtesy IBCAO (Jakobsson et al., 2008).

Figure 2. Hydrate saturation,  $S_H$ , within the 2-D system at t = 50, 100, 130, and 200 yr.

Figure 3. Gas saturation,  $S_G$ , within the 2-D system at t = 50, 100, 130, and 200 yr.

Figure 4. Flux of gaseous methane,  $Q_{CH4}$ , at the seafloor at t = 85, 100, 130, 160, and 200 yr.  $Q_{CH4}$  is presented here as mol CH<sub>4</sub> per m<sup>2</sup> at downslope position *x* for a 1 m-wide, 2-D slice of the continental slope.







