



## RESEARCH LETTER

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## Key Points:

- Structure II gas hydrates form below a bottom-simulating reflector due to thermogenic gas migration and coexist with free gas
- A chemical fractionation process provokes a transition at the bottom-simulating reflector from structure II to structure I hydrates
- Gas hydrate systems sourced by thermogenic fluids can be characterized by more hydrates below than above bottom-simulating reflectors

## Supporting Information:

- Supporting Information S1
- Table S1

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## Structure II gas hydrates found below the bottom-simulating reflector

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**Abstract** Gas hydrates are a major component in the organic carbon cycle. Their stability is controlled by temperature, pressure, water chemistry, and gas composition. The bottom-simulating reflector (BSR) is the primary seismic indicator of the base of hydrate stability in continental margins. Here we use seismic, well log, and core data from the convergent margin offshore NW Borneo to demonstrate that the BSR does not always represent the base of hydrate stability and can instead approximate the boundary between structure I hydrates above and structure II hydrates below. At this location, gas hydrate saturation below the BSR is higher than above and a process of chemical fractionation of the migrating free gas is responsible for the structure I-II transition. This research shows that in geological settings dominated by thermogenic gas migration, the hydrate stability zone may extend much deeper than suggested by the BSR.

## 1. Introduction

Natural gas hydrates, one of the most important organic carbon resources on Earth [Sloan and Koh, 2007; Boswell and Collett, 2011; Piñero et al., 2013], are mainly concentrated in deep water environments along continental margins and permafrost regions [Collett et al., 2009]. The stability of gas hydrates is controlled by four dominant factors: temperature, pressure, water salinity, and guest molecular composition. Excess gas saturation with respect to water is required for hydrates to form [Xu and Ruppel, 1999; Davie et al., 2004]. The impact of hydrate destabilization on climate change and its role as a future energy resource are widely debated [Collett et al., 2009; Boswell and Collett, 2011; Dickens, 2011; Ruppel, 2011; Hunter et al., 2013]. The origin of the gas-forming hydrates can be biogenic, thermogenic, or abiotic [Milkov, 2005; Collett et al., 2009; Johnson et al., 2015].

The crystalline structure of gas hydrates is controlled by gas composition. Structure I (S<sub>I</sub>) natural gas hydrates commonly include almost pure methane: this is the case for many microbially sourced gas hydrate systems [e.g., Milkov, 2005]. Structure II (S<sub>II</sub>) and H (S<sub>H</sub>) hydrates have a wider range of stability conditions compared to S<sub>I</sub> hydrates and can host a variety of gases, including methane and heavier order (C<sub>2+</sub>) hydrocarbons [Sassen and MacDonald, 1994; Milkov, 2005; Lu et al., 2007]. These gases are commonly thermogenic in origin and are sourced by relatively deep leaking reservoirs. Natural gas hydrate systems associated with thermogenic hydrocarbons have been observed in many different sedimentary basins worldwide [Diaconescu et al., 2001; Sassen et al., 2001a, 2001b, 2001c; Mazurenko et al., 2002; Blinova et al., 2003; Kida et al., 2006; Lu et al., 2007; Bourry et al., 2009; Pape et al., 2010, 2014; Ruffine et al., 2013; Smith et al., 2014; Serié et al., 2016]. However, S<sub>II</sub> and S<sub>H</sub> gas hydrates have often been directly sampled only at or close to the seafloor, without a complete penetration and direct sampling throughout the gas hydrate stability zone (GHSZ) [Sassen et al., 2001a, 2001b, 2001c; Pohlman et al., 2005; Lu et al., 2007; Bourry et al., 2009; Klapp et al., 2010]. Thus, predicting the vertical extent of the GHSZ and the hydrate distribution in these settings can be challenging [Boswell et al., 2012; Macelloni et al., 2015].

Historically, the bottom-simulating reflector (BSR) is the primary seismic indicator for the presence of a gas hydrate system and it is believed to closely approximate the base of the gas hydrate stability zone (BGHSZ) [Holbrook et al., 1996; Haacke et al., 2007; Hornbach et al., 2012]. However, major concerns have been raised regarding its unequivocal use in this respect, since gas hydrates have been observed to occur in areas lacking clear and continuous BSRs [Holbrook et al., 1996; Tsuji et al., 2009; Boswell et al., 2012]. The occurrence of multiple BSRs in some areas has led some authors to suggest that they could either (1) represent multiple BGHSZs for different hydrate structures or (2) indicate paleopressure and temperature conditions [Andreassen et al., 2000; Foucher et al., 2002; Bangs et al., 2005; Popescu et al., 2006; Pecher et al., 2014].

Furthermore, the presence of gas hydrates below a BSR has been inferred along the Cascadia Margin (site U1328, IODP 311), on the basis of a temperature and chlorinity anomaly measured just beneath the BSR [Riedel *et al.*, 2009] and on the Blake Ridge, because of the depth misfit between the predicted and the observed BSR, explained by some authors with capillary effects, ultimately provoking a zone of free gas and hydrate coexistence [Guerin *et al.*, 1999; Liu and Flemings, 2011].

This study is located on the middle-upper continental slope of Sabah. This area is dominated by a series of anticlines that formed in the past few million years as a result of far-field stress and gravitational tectonics [Ingram *et al.*, 2004; Hesse *et al.*, 2009; Morley, 2009]. Here oil and gas have been found within stacked siliciclastic turbiditic reservoirs, sourced by type III estuarine to continental Tertiary source rocks [Warren *et al.*, 2010].

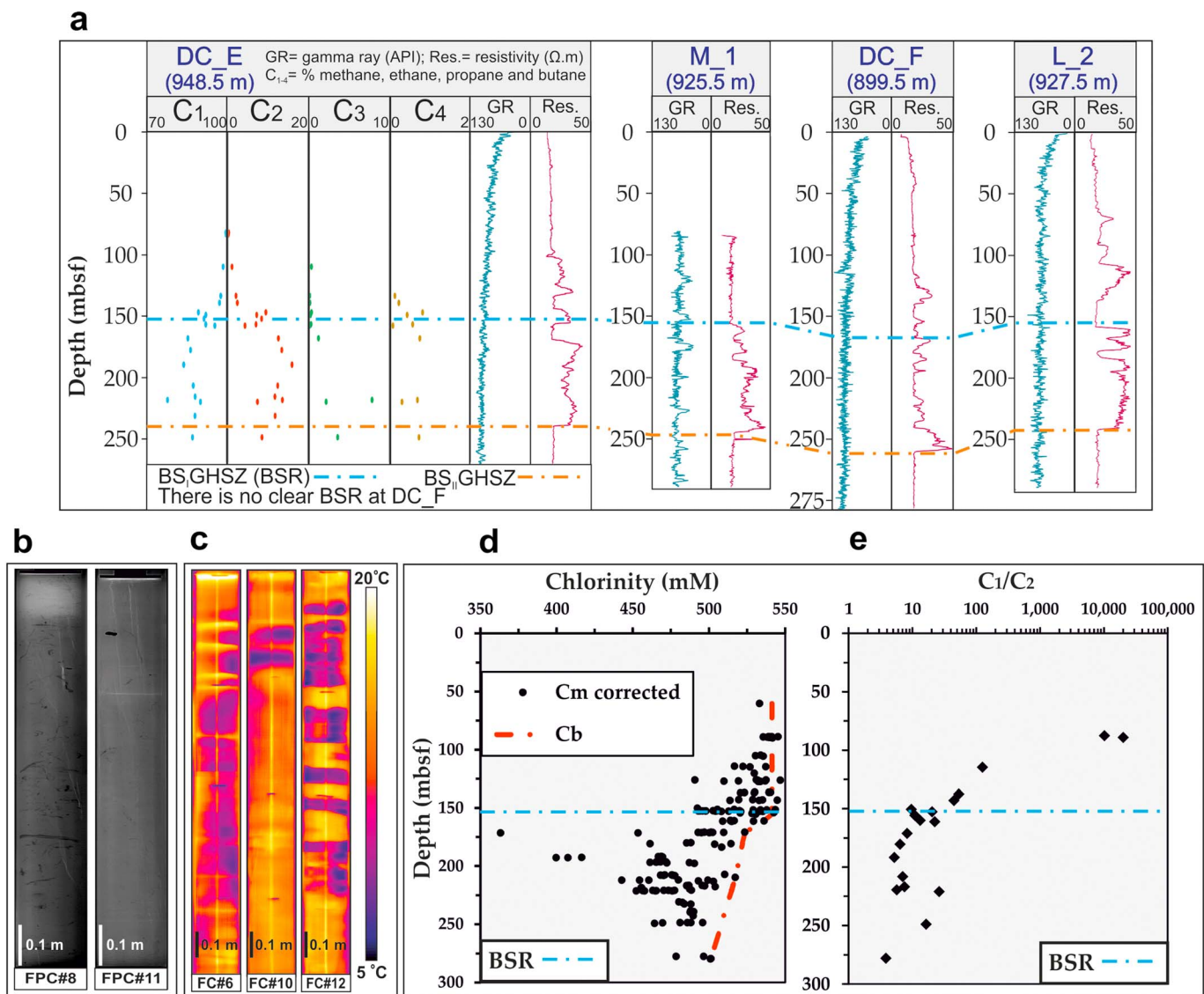
The presence of gas hydrates along this margin has been previously confirmed by the recognition of BSRs, as well as near-surface piston core sampling [Gee *et al.*, 2007; Warren *et al.*, 2010; Laird and Morley, 2011]. Here conventional thermogenic hydrocarbon traps are typically associated with gas hydrate systems, providing evidence of vertical leakage phenomena [Gee *et al.*, 2007; Warren *et al.*, 2010]. Specifically, this research is focused on the gas hydrate system overlying the Gumusut-Kakap oil and gas reservoir, discovered by Shell in 2003 [Hadley *et al.*, 2008]. The preliminary report of Hadley *et al.* [2008] was primarily aimed at a shallow geohazard study. However, these authors noted the presence of  $S_{II}$  hydrates below a BSR and  $S_I$  hydrates above it in a single borehole from the analysis of core data and resistivity measurements. This paper builds on their initial analyses by looking at three other wells to (1) further constrain the presence of gas hydrates below a BSR, (2) estimate their saturation, and (3) calibrate gas hydrate indicators with stability modeling.

## 2. Data and Methods

This research integrates a 3-D seismic survey with well log and core data. The 3-D seismic survey covers an area of 37.32 km<sup>2</sup> (Figure 1a), with a 6.25 m bin spacing, 1 ms sampling interval, and a 25–150 Hz frequency band at –20 dB for the first 1750 ms TWT (Two-Way Time). The average vertical resolution is 5.25 m [Widess, 1973]. Logging while drilling (LWD) data were acquired using the Halliburton downhole logging tools from four closely spaced boreholes (named DC\_E, M\_1, DC\_F, and L\_2) (Figure 1a). Conventional well log data acquisition included gamma ray, resistivity, density, and neutron porosity measurements. In particular, resistivity, which is diagnostic to identify hydrate-bearing sediments [e.g., Cook *et al.*, 2010], was measured with the EWR<sup>®</sup>-Phase 4 multiaarray propagation tool, which also allowed the acquisition of horizontal ( $R_h$ ) and vertical ( $R_v$ ) resistivity data. Resistivity images were acquired at DC\_E using the Simultaneous Acoustic and Resistivity Imager (STAR) device, from ~55 to ~285 m below seafloor (mbsf).  $P$  and  $S$  wave velocities were acquired at DC\_E during the wireline acquisition with the Cross-Multipole Array Acoustic log from Baker Hughes. A synthetic seismogram was built to calibrate the seismic with well data by using the wireline velocity and LWD density logs acquired at DC\_E and applying a Ricker wavelet in line with the 3-D seismic data.

The modern standard techniques to recover conventional and pressure cores and analyze natural gas hydrate-bearing sediments [cf. Tréhu *et al.*, 2004; Weinberger *et al.*, 2005; Schultheiss *et al.*, 2006; Lee *et al.*, 2013; Ryu *et al.*, 2013; Holland and Schultheiss, 2014] were applied at DC\_E. A total of 14 conventional (i.e., nonpressurized) cores, having an average length of 166.5 cm, were successfully recovered with the hydraulic percussion Fugro Corer (FC). Infrared (IR) images and temperature data were acquired in the FC cores to identify thermal anomalies from dissociating hydrates. Additional data were obtained using the WISON/WIP XP push sampler [cf. Hawkins and Narkus, 1998], which allowed the recovery of nonpressurized samples using either a PVC liner or a thin-walled Shelby tube. All the recovered FC cores, liner samples, and Shelby tubes were used for pore water chlorinity measurements.

The Fugro Pressure Corer (FPC) and the Hyace Rotary Corer (HRC) allowed the recovery of 1 m length pressure cores under hydrate stability conditions. Once successfully recovered, the core was subjected to nondestructive measurements before being depressurized. A total of 20 pressure cores (13 FPC and 7 HRC cores) have been recovered. All 13 FPC cores were used for pore water chlorinity measurements, whereas only the eight FPC and four HRC cores successfully recovered were used for gas geochemistry. An empirical baseline chlorinity ( $C_b$ ) profile was built following Ussler and Paull [2001] and represents the plausible pore water conditions before freshening due to gas hydrate dissociation. Gamma density (error  $\pm 2\%$ ),  $P$  wave velocity (error  $\pm 1\%$ ), and X-ray images (pixel resolution of  $\sim 120 \mu\text{m}$ ) of successfully recovered FPC cores, measured in the Geotek Multi Sensor



**Figure 2.** Well log and core data providing evidence of gas hydrates below the BSR. (a) Phase shift deep smoothed resistivity (Res.) and gamma ray (GR) data for wells DC\_E, M\_1, DC\_F, and L\_2. The water depths of the four wells are indicated in brackets. Bulk gas composition data from cores retrieved at DC\_E are also included. Note the increase of C<sub>2+</sub> hydrocarbons below the BSR (i.e., the BS<sub>I</sub>GHSZ), indicated with a blue dotted line. The interpreted BS<sub>II</sub>GHSZ is marked with an orange dotted line. (b) X-ray images of FPC cores at DC\_E. FPC#8 and FPC#11 have been respectively retrieved approximately at the BSR depth and ~70 m below. (c) IR images of FC cores. FC#6, FC#10, and FC#12 have been respectively retrieved ~40 m, ~60 m, and ~70 m below the BSR. (d) Chlorinity profile at DC\_E showing the measured values (C<sub>m</sub>) and the estimated baseline profile (C<sub>b</sub>). (e) Methane/ethane (C<sub>1</sub>/C<sub>2</sub>) ratio measured in cores at DC\_E.

horizontal and vertical resistivity (Figures S5 and S6 in the supporting information) [Cook *et al.*, 2010]. Nevertheless, we do not exclude the presence of gas hydrates within the primary pore space because such occurrence is not readily observable in X-ray images [Holland *et al.*, 2008].

At well DC\_E, the estimated *Sh* from both core and well log data agree. Our estimates using resistivity data in the four wells indicate that *Sh* is consistently higher below than above the BSR. *Sh* above the BSR is typically 1–5% of pore space but is 5–20% of pore space below, although it locally exceeds 20–30% of the pore space (Figure 3). There is no clear correlation between wireline velocity logs at DC\_E and the estimated gas hydrate saturations apart from a strong positive peak in *S* wave velocity just above the BSR which could link with gas hydrates that have considerably increased the sediment stiffness [Riedel *et al.*, 2014].

Unfortunately, the seismic data below the BSR are severely attenuated and it is not possible to confidently calibrate the abrupt resistivity reduction, occurring at 237–260 m, with any seismic reflection. However, the

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