



## Research paper

# Gas hydrate distribution and carbon sequestration through coupled microbial methanogenesis and silicate weathering in the Krishna–Godavari Basin, offshore India



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## ABSTRACT

The National Gas Hydrate Program Expedition 01 cored ten sites in the Krishna–Godavari basin, located on the southeastern margin of India. A comprehensive suite of pore water solute concentrations and isotope ratios were analyzed to investigate the distribution and concentration of gas hydrate along the margin, *in situ* diagenetic and metabolic reactions, and fluid migration and flow pathways. Gas hydrate was present at all of the sites cored, and *in situ* microbial methanogenesis leads to estimates of depth-integrated average gas hydrate saturations that are typically <5%. Deep-sourced fluid and gas migration produces gas hydrate saturations up to 68% along an isolated coarser-grained stratigraphic horizon at Site 15 and up to 41% within a fractured clay-dominated system at Site 10. Our results show that the CO<sub>2</sub> produced through net microbial methanogenesis is effectively neutralized by silicate weathering throughout the sediment column drilled at each site (~100–300 m), buffering the pore water pH and generating excess alkalinity via the same reaction sequence as continental silicate weathering. Most of the excess alkalinity produced by silicate weathering in the Krishna–Godavari basin is sequestered in Ca- and Fe-carbonates as a result of ubiquitous calcium release from weathering detrital silicates and dissolved Fe production within the methanogenic sediments. Formation of secondary hydrous silicates (e.g. smectite) related to incongruent primary silicate dissolution acts as a significant sink for pore water Mg, K, Li, Rb, and B. The consumption of methane through anaerobic oxidation of methane, sequestration of methane in gas hydrate, and sequestration of dissolved inorganic carbon in authigenic carbonates keeps methanogenesis as a thermodynamically feasible catabolic pathway. Our results combined with previous indications of silicate weathering in anoxic sediments in the Sea of Okhotsk, suggest that silicate weathering coupled to microbial methanogenesis should be occurring in continental margins worldwide, providing a net sink of atmospheric CO<sub>2</sub>.

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## 1. Introduction

Elevated primary production, the export of terrestrial organic matter from land to the ocean, and high sedimentation rates at continental margins contribute to an increased burial flux of metabolizable organic matter at the seafloor (e.g. Hedger and

Keil, 1995; Gélinas et al., 2001). As a result, seafloor microbial metabolism causes rapid pore water anoxia and drives high rates of sulfate reduction, providing biogeochemical conditions that promote microbial methanogenesis at shallow depths in the sediment column (e.g. Claypool and Kaplan, 1974; Froelich et al., 1979). Microbial methanogenesis is ubiquitous along continental margins worldwide, and within the gas hydrate stability zone (typically water depths >500 m; >300 m in the Arctic), gas hydrates form when the concentration of methane dissolved in the pore water exceeds gas hydrate solubility.

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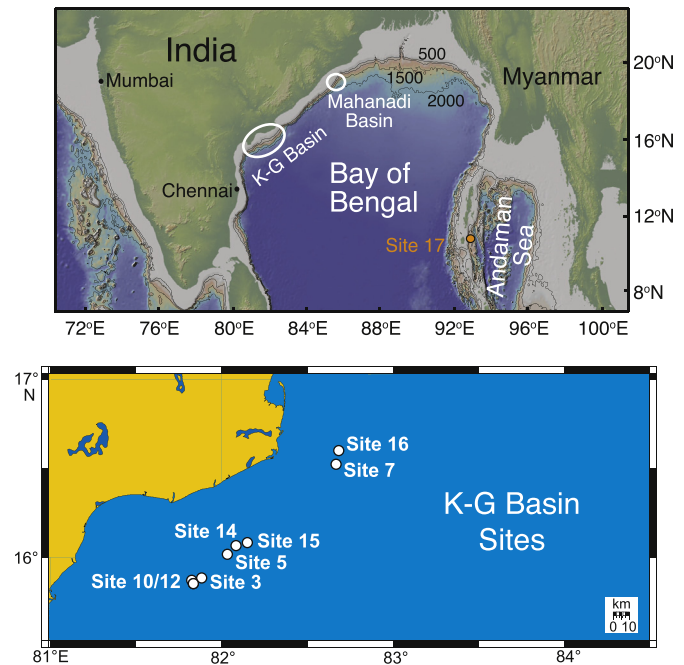
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Estimates of the global rate of microbial methanogenesis in marine sediments ranges from ~2 to 20 Tmol CH<sub>4</sub>/yr (Hinrichs and Boetius, 2002; Reeburgh et al., 1993; Wallmann et al., 2012). Most of the methane that is produced in continental margin sediments is either sequestered in gas hydrate or is oxidized through anaerobic oxidation of methane (AOM) at the sulfate–methane transition zone (SMTZ). An unconstrained fraction of the methane produced bypasses these filters and is emitted to the ocean. The cycling of methane in marine sediments has been actively studied over the past several decades, but far less attention has been paid to the cycling of CO<sub>2</sub> in methanogenic sediments.

During net microbial methanogenesis, CO<sub>2</sub> is also produced such that for every mole of CH<sub>4</sub> released to the pore water, approximately a mole of CO<sub>2</sub> is released. Recent studies based on piston cores (<25 m in length) in the Sea of Okhotsk and Black Sea (Aloisi et al., 2004; Wallmann et al., 2008) suggest that much of the CO<sub>2</sub> produced in shallow sediments through microbial methanogenesis is neutralized through weathering of detrital silicate minerals leading to the production of alkalinity. It is not known whether this is ubiquitous within gas hydrate-bearing sediments worldwide, but it may be an important sink of CO<sub>2</sub> and source of alkalinity to the ocean (Wallmann et al., 2008). Additionally, the removal of dissolved CO<sub>2</sub> may influence the thermodynamics and rate of methanogenesis.

Recent estimates of the global gas hydrate inventory, based on multiple approaches, suggest that ~400–3000 Gt of methane-bound carbon is stored in gas hydrate in marine sediments (e.g. Archer et al., 2009; Buffet and Archer, 2004; Miliukov, 2004; Piñero et al., 2013). Considering the large mass of methane carbon stored in the global gas hydrate reservoir, it may be a potential energy resource (e.g. Boswell and Collett, 2011). Since the gas hydrate reservoir is sensitive to pressure and temperature perturbations, it also has the potential to affect climate (e.g. Dickens et al., 1995; Kvenvolden, 1993). Gas hydrates, however, are typically not uniformly distributed in the sediment within the gas hydrate stability zone (e.g. Malinverno, 2011; Torres et al., 2008). This complicates assessing their global inventory and resource potential as well as predicting the vulnerability and environmental consequences of gas hydrate dissociation driven by environmental change. The Indian National Gas Hydrate Program (NGHP) Expedition 01 was designed to study the occurrence of gas hydrate off the Indian Peninsula and along the Andaman convergent margin (Fig. 1) with emphasis on understanding the geologic and geochemical controls on methane dynamics and the distribution of gas hydrate in the two settings (Collett et al., 2008).

Here we report on geochemical analyses of pore water samples collected at high-resolution from ten sites cored in the Krishna–Godavari (K–G) basin during NHGP Expedition 01 (Supp. Table 1) and focus our discussion on six of these sites (Table 1; Fig. 1). A comprehensive suite of major, minor, and trace element concentrations, as well as isotope ratios, were analyzed to investigate the spatial distribution of gas hydrates across the margin, *in situ* diagenetic and metabolic reactions, fluid migration and flow pathways, and fluid and gas sources. Here we focus on both CH<sub>4</sub> and CO<sub>2</sub> dynamics in the sediment column to better understand the processes and sinks controlling CO<sub>2</sub> produced through methanogenesis. The pore water geochemical data set presented herein is one of the most extensive collected to date from a gas hydrate system, and provides new insights into co-occurring microbial and inorganic reactions that control carbon and elemental fluxes. These results highlight the importance of silicate weathering driven by microbial methanogenesis in generating excess alkalinity that leads to the long-term storage of carbon through pervasive authigenic carbonate precipitation in continental margin sediments.



**Figure 1.** Top. Bathymetric map showing the locations of the Krishna–Godavari and Mahanadi basins and Site 17 cored in the Andaman Sea during NGHP Expedition 01. Bottom. Map of the sites cored in the K–G basin. Adapted from Collett et al. (2008).

## 2. Geologic setting

The eastern continental margin of India evolved as a consequence of rifting between India and Australia/Antarctica in the Late Jurassic and Early Cretaceous. The Krishna–Godavari basin (K–G basin), located along the central east coast of India, occupies an onshore area of ~28,000 km<sup>2</sup> and extends beyond the mid-slope, encompassing an offshore area of 24,000–49,000 km<sup>2</sup> (Radhakrishna et al., 2012). The tectonic activity along the eastern continental margin of India resulted in the formation of several NE–SW trending horst and graben-like structures (Rao, 2001), and the grabens are filled with a thick pile of sediments with a maximum sediment thickness of >7 km (Bastia and Nayak, 2006). The Godavari and Krishna rivers supply the bulk of the detrital sediments to the K–G basin.

The collision between the Indian and Eurasian plates resulted in the building of the Himalayas. Subsequent monsoon activity promoted rejuvenation of major rivers (Ganges, Brahmaputra, Mahanadi, Krishna, Godavari, and Cauvery) and minor tributaries, which collectively transport large amounts of sediments into the Bay of Bengal (Curry and Moore, 1971). This sediment discharge is manifested in the forms of deltas and delta fronts on the inner shelf, extending beyond the shelf break (Ramana et al., 2009). The shelf is incised by headward eroding point-source canyons that are now abandoned and filled with clay and reworked sediments. Several V-shaped canyons flanked by steep faults characterize the upper to mid-slope regions, while turbidity channels and levee wedges transverse the shelf and slope (Ramana et al., 2009; Riedel et al., 2011). Widespread mass transport deposits have been observed over the modern upper continental slope and have also contributed to the total sediment accumulation/dynamics (Ramprasad et al., 2011; Shanmugam et al., 2009; Hong et al., 2014). The high sedimentation rates on the K–G basin slope and POC concentrations of 1.5–2.5 wt.% create favorable conditions for methane generation. Indeed, the K–G basin is one of the most important petroliferous basins of India, and large conventional gas and gas hydrate deposits

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