



Methane in shallow subsurface sediments at the landward limit of the gas hydrate stability zone offshore western Svalbard

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Abstract

Offshore western Svalbard plumes of gas bubbles rise from the seafloor at the landward limit of the gas hydrate stability zone (LLGHSZ; ~400 m water depth). It is hypothesized that this methane may, in part, come from dissociation of gas hydrate in the underlying sediments in response to recent warming of ocean bottom waters. To evaluate the potential role of gas hydrate in the supply of methane to the shallow subsurface sediments, and the role of anaerobic oxidation in regulating methane fluxes across the sediment–seawater interface, we have characterised the chemical and isotopic compositions of the gases and sediment pore waters. The molecular and isotopic signatures of gas in the bubble plumes ($C_1/C_{2+} = 1 \times 10^4$; $\delta^{13}C\text{-CH}_4 = -55$ to -51‰ ; $\delta D\text{-CH}_4 = -187$ to -184‰) are similar to gas hydrate recovered from within sediments ~30 km away from the LLGHSZ. Modelling of pore water sulphate profiles indicates that subsurface methane fluxes are largely at steady state in the vicinity of the LLGHSZ, providing no evidence for any recent change in methane supply due to gas hydrate dissociation. However, at greater water depths, within the GHSZ, there is some evidence that the supply of methane to the shallow sediments has recently increased, which is consistent with downslope retreat of the GHSZ due to bottom water warming although other explanations are possible. We estimate that the upward diffusive methane flux into shallow subsurface sediments close to the LLGHSZ is $30,550 \text{ mmol m}^{-2} \text{ yr}^{-1}$, but it is $<20 \text{ mmol m}^{-2} \text{ yr}^{-1}$ in sediments further away from the seafloor bubble plumes. While anaerobic oxidation within the sediments prevents significant transport of dissolved methane into ocean bottom waters this amounts to less than 10% of the total methane flux (dissolved + gas) into the shallow subsurface sediments, most of which escapes AOM as it is transported in the gas phase.

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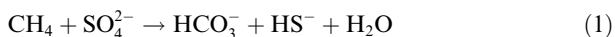
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1. INTRODUCTION

Methane (CH₄) is a potent greenhouse gas, and its atmospheric concentration has increased by more than 10% over the past three decades (Nisbet et al., 2014). Atmospheric methane concentrations are closely correlated with temperature over glacial-interglacial cycles (e.g., Möller et al., 2013), and methane emissions may have played a key role in major climate excursions in the past, including the Paleocene-Eocene thermal maximum (e.g., Dickens, 2011; Bowen et al., 2014). Many natural sources of methane are sensitive to climatic changes including wetlands, permafrost, and methane hydrate in terrestrial and marine environments (Ciais et al., 2013). Characterization of emissions from these sources is paramount to the assessment of how levels of atmospheric methane will evolve in the context of future global climate.

Methane in marine sediments is produced by microbial and thermal degradation of organic carbon. If sediment pore waters become methane saturated, and temperature is low and pressure relatively high, then methane hydrate, an ice-like solid in which gas molecules are trapped in cages of water molecules, may form (e.g., Kvenvolden, 1993; Hester and Brewer, 2009). Methane hydrate is stable on the continental margin at water depths >200 m at high latitudes (or shallower in some permafrost settings; Krey et al., 2009) and on a global scale may contain >500 Gt carbon, although this reservoir remains poorly quantified (e.g., Milkov, 2004; Archer, 2007; Wallmann et al., 2012; Piñero et al., 2013).

A key uncertainty for climate models is the proportion of methane produced in deep marine sediments that escapes to reach the sediment–seawater interface and potentially the atmosphere. Methane rises from depth through permeable sediment strata, faults, fractures and cracks in solution, and as gas if sediment pore waters are supersaturated (Judd and Hovland, 2007). In near-surface sediments where sulphate is present, dissolved methane can be oxidised by a consortium of archaea and sulphate-reducing bacteria. This process is known as anaerobic oxidation of methane (AOM; Eq. (1); Boetius et al., 2000):



The sub-seafloor depth interval in which methane and sulphate are consumed by AOM and concentrations of both species are nearly depleted is known as the sulphate-methane transition zone (SMTZ). The depth of the SMTZ is a qualitative proxy for the upwards methane flux into shallow sediments, as the availability of sulphate is limited by its rate of diffusion from seawater such that rapid sulphate consumption pushes the SMTZ closer to the seafloor in regions of high methane supply (e.g., Borowski et al., 1996). Hydrogen carbonate (HCO₃⁻) ions produced by AOM react with calcium ions present in sediment pore waters to form calcium carbonate (Sun and Turchyn, 2014) (Eq. (2)):



However, if the methane flux is sufficient to overwhelm the oxidising capacity of the microbial consortia (Archer

et al., 2009), or the re-supply of sulphate from seawater by diffusion (Knittel and Boetius, 2009), methane is released into the water column and potentially into the atmosphere. Methane in the gas phase is not available to microbes and therefore escapes oxidation.

As the Earth and its oceans warm, there is concern that gas hydrate in marine sediments will be destabilized, releasing methane that will reinforce the greenhouse effect (e.g., Krey et al., 2009). This is supported by observations of methane bubble plumes in the water column near the landward limit of the gas hydrate stability zone (LLGHSZ), rising from sediments in which gas hydrate may have recently destabilized (e.g., Westbrook et al., 2009 (Arctic); Hautala et al., 2014 (North Pacific); Skarke et al., 2014 (North Atlantic)). Arctic regions, where climate warming is amplified (e.g. Parmentier et al., 2013), and cold bottom waters allow hydrate to form in shallow sediments where it is more susceptible to warming (e.g. Kretschmer et al., 2015), are of particular interest.

More than 200 methane bubble plumes were discovered rising from the seabed at water depths close to the LLGHZ offshore western Svalbard (Westbrook et al., 2009). In this study, we characterise the chemical composition of gases in sediments from the vicinity of the LLGHSZ offshore Svalbard, and assess the spatial distribution and temporal variability of methane fluxes into the shallow subsurface, and across the sediment–seawater interface. We examine the geochemical evidence that hydrate dissociation fuels high methane fluxes into shallow sediments, and the role of AOM in regulating methane release from the seafloor.

2. STUDY AREA AND SAMPLING

The continental margin offshore western Svalbard (Fig. 1) was shaped by the Pleistocene-Pliocene advance and retreat of the Svalbard-Barents Sea ice sheet (Sarkar et al., 2012). Glacial ice withdrew from the continental shelf about 13 thousand years ago (Elverhøi et al., 1995; Jessen et al., 2010). On the shelf and upper slope, patchy glacial sediments overlie a sequence of seaward-dipping marine sediments (Rajan et al., 2012; Sarkar et al., 2012). At water depths of more than 700 m, sediments are underlain by extensive gas hydrate deposits (Eiken and Hinz, 1993; Vogt et al., 1999; Carcione et al., 2005; Vanneste et al., 2005; Fisher et al., 2011; Sarkar et al., 2011; Bünz et al., 2012; Smith et al., 2014; Plaza-Faverola et al., 2015) that likely extend upslope to the limit of hydrate stability, although at lower hydrate concentrations (e.g., Chabert et al., 2011).

The present-day landward limit (LL) of the GHSZ (LLGHSZ) at the seabed, i.e. the shallowest water depth beneath which hydrate is stable, is defined by water depth (pressure) and the temperature of overlying bottom water of the West Spitsbergen Current (WSC) which flows northward along the upper slope. Seasonal variations in bottom water temperature are ~1.5 °C, which results in seasonal shifts in the LLGHSZ between ~360 m water depth and ~410 m water depth (Berndt et al., 2014). Critically, the WSC appears to have warmed over the last ~60 years (Westbrook et al., 2009; Ferré et al., 2012), and the present

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