



The challenges of quantifying the carbon stored in Arctic marine gas hydrate



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ABSTRACT

The quantification of the carbon stored in gas hydrate (GH) bearing marine sediments still remains a challenge. Despite recent efforts to develop approaches to better estimate the GH inventory globally, these estimates are still highly unconstrained due to insufficient field data and poor understanding of the mechanisms fuelling the GH stability zone (GHSZ). Here we use geophysically-derived GH saturations to constraint estimates of model-derived Arctic marine GH inventory at present. We also estimate the potential carbon released from GH dissociation under a seabed warming of 2 °C over 100 yr. We estimate an inventory ranging between 0.28 and 541 Gt of carbon, which upper bound results in average GH saturations of 0.25%. Our upper bound is mainly controlled by our imposed upwards carbon-rich fluid flow of 0.01 cm yr⁻¹ and it is five times greater than the most recent estimate that only considers in-situ degradation of particulate organic carbon (POC). To obtain the seismically-inferred GH saturations of 5–10% offshore west of Svalbard and in the Beaufort Sea, an upwards advection of carbon-rich fluids equivalent to 0.02–0.04 cm yr⁻¹ is required. This mechanism may be the most important source of carbon reaching the GHSZ in Arctic marine sediments. A 2 °C seabed temperature increase over 100 yr may reduce the GH inventory by about 88.44% (0.7 Gt C) if POC is the only source, and by about 5.4% (29.7 Gt C) if the main source of carbon is the upwards advection of carbon-rich fluids.

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1. Introduction

At present, various countries (USA, Canada, Japan, India, China, South Korea) have important R&D programs to make gas hydrate (GH) exploitation economically feasible in the relatively near future. Therefore, the first step to understand the potentiality of hydrate as an energy resource or as a future impact to the climate is to quantify its inventory, which still is highly uncertain.

Recent global estimates of the total carbon stored in GH bearing sediments range between ~500 and 3000 Gt of which 116 Gt may be stored in the Arctic (e.g., Kretschmer et al., 2015). Hydrates are most sensitive to ocean warming at high latitudes and in shallow water depths (e.g., Hunter et al., 2013), and for a 100 yr warming period the Arctic presents the maximum absolute methane released from hydrate dissociation with a global contribution of

39% (140 ± 10 Mt C; Kretschmer et al., 2015). However, Kretschmer et al. (2015) do not consider the upward advection of deep methane-rich fluids into the GH stability zone (GHSZ) from processes other than mechanical compaction, such as dewatering, which may significantly increase the present day GH inventory and associated future methane release. Besides, the transformation of their estimates into GH saturation results in much smaller saturations than those inferred from seismic and controlled sourced electromagnetic (CSEM) data in several Arctic locations (e.g., Andreassen et al., 1997; Chabert et al., 2011; Goswami et al., 2015).

Uncertainties in the parameters controlling the thickness of the GHSZ (pressure, seabed temperature, geothermal gradient, salinity and phase boundary) and in the type of carbon sources and amount of carbon reaching the GHSZ result in a large range of possible estimates of the total carbon stored in GH. Here, we illustrate the influence of uncertainties in the parameters controlling the thickness of the GHSZ by considering a rather large perturbation of ±30% in the calculation of the Arctic marine GH inventory from published state-of-the-art transfer functions (Wallmann et al., 2012; Piñero et al., 2013). We also present an analysis where some of these

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estimates are constrained with GH saturations derived from geophysical data and propose an explicit function that allows the estimation of an equivalent upward fluid flow of methane-rich fluids into the GHSZ required satisfying geophysically-derived saturations. This function can be used anywhere when the accumulation of particulate organic carbon is not sufficient to explain average hydrate saturations above 1%. We finally assess the potential GH-derived carbon that could be released under a 2 °C seabed warming scenario over 100 yr for different present-day Arctic GH inventories.

2. Methodology

The carbon stored in Arctic marine GHs was calculated using the transfer functions proposed by Wallmann et al. (2012) for diffusive-controlled geological systems, and for fully compacted and steady state compacted sediments. To consider other possible sources of dissolved methane into the GHSZ, Piñero et al. (2013) transfer function (Eq. (1)) was also applied. These transfer functions are fitting equations to the numerical results from a reactive transport code that considers the dominant physical and biogeochemical processes and parameters including: sediment compaction, the solubility of methane in pore water, the formation and dissociation of GH and formation and dissolution of free methane gas in pore water, diffusive and advective transport of dissolved constituents, input and degradation of particulate organic carbon (POC) and particulate organic nitrogen (PON) via sulfate reduction and methanogenesis, anaerobic oxidation of methane (AOM), and formation and adsorption of ammonium, dissolved inorganic carbon (DIC) and methane (Piñero et al., 2013). The input parameters for the transfer functions are: (i) thickness GHSZ (H_{GHSZ} , m), (ii) sedimentation rate (SR, cm kyr^{-1}), (iii) POC (wt %), and (iv) upward advective fluid flow from mechanisms other than mechanical compaction (FF, cm yr^{-1}).

$$m_c = \sum_n^N \left\{ \begin{array}{l} \left[\left(c_1 \cdot H_{\text{GHSZ}}^2 (c_3 - \frac{1}{\text{SR}}) \cdot (\text{POC} + c_4 \cdot \text{FF}^{c_5}) \text{POC}^{c_6} \right) \cdot A \right]; \quad \text{FF} \geq 0.0001 \text{SR} (2 + \ln[\text{POC}]) \\ \left[\left(m_c^* \cdot c_7 \cdot 10^{-8} H_{\text{GHSZ}}^{c_8} (1 - \frac{1}{\text{SR}}) \text{FF} \cdot \text{POC}^{c_9} \cdot A \right) \right]; \quad \text{FF} < 0.0001 \text{SR} (2 + \ln[\text{POC}]) \end{array} \right\}_n \quad (1)$$

In Eq. (1) m_c (kg) is the total carbon locked in GH, N is the number of model cells, m_c^* (kg m^{-2}) is the carbon locked in GH per m^2 of seabed area calculated using Wallmann et al. (2012) transfer function for steady-state compaction, A (m^2) is the seabed area, and the fitting coefficients are: [$c_1 = 0.024$; $c_2 = 1.587$; $c_3 = 0.0224$; $c_4 = 266084$; $c_5 = 2.75$; $c_6 = 0.063$; $c_7 = 0.003$; $c_8 = 4.68$; $c_9 = 2.31$]. Please note that the ascent of free methane gas, which may be another source for methane in the GHSZ, is not considered in these functions.

2.1. Volume of the GHSZ

To calculate the present-day volume of the marine GHSZ in the Arctic under steady state conditions, bathymetry, seabed temperature and geothermal gradient data were collected (Fig. 1), and water salinity and gas composition were assumed. The bathymetric data was obtained from The International Bathymetric Chart of the Arctic Ocean (IBCAO) project (<http://www.ngdc.noaa.gov/mgg/bathymetry/arctic/downloads.html>), the seabed temperature data

from The National Oceanographic Data Centre website (<http://www.nodc.noaa.gov/cgi-bin/OC5/WOA09/woa09.pl>), and the geothermal gradient data from The Global Heat Flow Database of the International Heat Flow Commission (<http://www.heatflow.und.edu/index2.html>). Note that, since we directly use geothermal gradient data, we do not need to assume any thermal conductivity value, which is normally an uncertainty source (e.g. Burwicz et al., 2011; Piñero et al., 2013). A value of 3.5 wt% Arctic Ocean salinity (Talley et al., 2011) and Structure I pure methane hydrate were assumed, the later based on other hydrate-related studies in the Arctic (e.g., Marín-Moreno et al., 2015) and because methane hydrate makes the 80% of the total inventory of naturally occurring GHs (Kvenvolden et al., 1993). We consider a model resolution of $2500 \times 2500 \text{ m}^2$ and the above datasets were interpolated and extrapolated to that resolution. In each model cell, the thickness of the GHSZ was given by the distance between the seabed and the intersection of the cell's temperature structure (obtained using the cell's seabed temperature and geothermal gradient) with six different methane hydrate phase boundaries: (1) and (2) Dickens and Quinby-Hunt (1994, 1997), (3) Distribution Coefficient Method or K_{vis} -Method (Sloan and Koh, 2008), (4) Moridis et al. (2008), (5) Tishchenko et al. (2005) and (6) Lu and Sultan (2008). Water depth was converted to hydrostatic pressure assuming a constant water density of 1046 kg m^{-3} (Giustiniani et al., 2013). Sloan and Koh's (2008) and Moridis' (2008) curves are defined for pure water and Dickens and Quinby-Hunt (1994) for 3.35 wt% salinity. These GH stability curves were converted to 3.5 wt% salinity curves using the relationship from Dickens and Quinby-Hunt (1997). For the conversion, we assumed a pure water fusion temperature of 273.2 K, a pure water fusion enthalpy of 6008 J mol^{-1} , an enthalpy of GH dissociation of 54200 J mol^{-1} , a hydration number of 6 ($\text{CH}_4 \cdot 6\text{H}_2\text{O}$), and Blangden's law (Ladd, 1998) to calculate the fusion temperature of water in an electrolyte solution of 3.5 wt% salinity. For Blangden's law, a water cryo-

scopic constant of 1853 Kg mol^{-1} and a NaCl van't Hoff factor of 2 were considered. The average thickness of the GHSZ in each model cell (Fig. 2A) was calculated using the different phase boundaries within their valid range of application (Table 1).

2.2. Sedimentation rate and particulate organic carbon

Two different methods were considered to calculate the sedimentation rate. Method 1 uses the water depth vs sedimentation rate relationship for Holocene sediments from Burwicz et al. (2011), and Method 2 uses an average sedimentation rate from the ratio between the sediment thickness (Whittaker et al., 2013) and the age of oceanic crust (Müller et al., 2008). The data for Method 2 were obtained from The National Oceanographic Data Centre website (<http://www.ngdc.noaa.gov/mgg/sedthick/>; http://www.ngdc.noaa.gov/mgg/ocean_age/ocean_age_2008.html), and in the cells where the data were not available, and could not be reliably extrapolated, the sedimentation rate calculated with Method 1 was used. GH would have formed over a period much larger than the

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