



# Dissolved methane concentrations in the water column and surface sediments of Hanna Shoal and Barrow Canyon, Northern Chukchi Sea



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## ARTICLE INFO

### Keywords:

Methane  
Arctic zone  
Chukchi Sea  
Hanna Shoal  
COMIDA

## ABSTRACT

Current estimates of methane (CH<sub>4</sub>) flux suggest that Arctic shelves may be a significant source of atmospheric CH<sub>4</sub>, a potent greenhouse gas. However, little information is known about the CH<sub>4</sub> flux from most Arctic shelves, other than the East Siberian Arctic Shelf. We report here dissolved CH<sub>4</sub> concentrations in the water column and within surface sediments of the Northern Chukchi Sea. We hypothesized that this area contains high concentrations of CH<sub>4</sub> because it receives nutrient rich waters through the Bering Strait, promoting primary production that enhances an organic-rich material flux to the seafloor and eventual microbial methanogenesis in the sediments. In August 2012, as part of the Chukchi Sea Offshore Monitoring in Drilling Area (COMIDA) project, fourteen stations were sampled on Hanna Shoal, a shallow feature on the shelf, and ten stations across the undersea Barrow Canyon. On Hanna Shoal, water column CH<sub>4</sub> concentrations ranged from 14 to 74 nM, and surface concentrations were up to 15 times supersaturated in CH<sub>4</sub> compared to equilibrium with the average atmospheric concentrations (3 nM). CH<sub>4</sub> concentrations at the sediment-water interface were around 1,500 nM, and typically increased with depth in the sediment. At the head of Barrow Canyon, water column CH<sub>4</sub> concentrations ranged from 5 to 46 nM, with the highest concentrations in the deepest waters that were sampled (118 m). Overall, the calculated fluxes to the atmosphere ranged from 1 to 80 μmol CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> for Hanna Shoal and 4 to 17 μmol CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> across the Barrow Canyon stations. Although there was a large range in these fluxes, the average atmospheric flux (20 μmol CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) across Hanna Shoal was 12 times lower than the flux reported from the East Siberian Arctic Shelf in summer. We conclude that while there is a positive flux of CH<sub>4</sub> to the atmosphere, this part of the Chukchi Sea is not a significant source of atmospheric CH<sub>4</sub> compared to the East Siberian Sea shelf.

## 1. Introduction

### 1.1. Methane

Methane (CH<sub>4</sub>) concentrations in the atmosphere have been increasing over the last century (Forster et al., 2007). As a potent greenhouse gas, increasing CH<sub>4</sub> concentrations will provide a positive feedback to climate warming. While the majority of atmospheric CH<sub>4</sub>, ~485 Tg y<sup>-1</sup>, originates from terrestrial sources (i.e. wetlands, biomass burning, ruminant animals, etc.), oceans and freshwater systems were estimated to contribute ~10 Tg y<sup>-1</sup> or 2% of the global CH<sub>4</sub> sources (Reeburgh, 2007). This value continues to be reevaluated and has recently been set as high as 100 Tg y<sup>-1</sup> due to freshwater inputs

(Bastviken et al., 2011). Marine regions at particularly high latitude could conceivably increase in importance as a source because of the presence of CH<sub>4</sub> hydrates in permafrost, including undersea permafrost, which is susceptible to release as the climate warms. Recent isotopic evidence moreover suggests that increased CH<sub>4</sub> fluxes are due to degrading permafrost in Arctic tundra settings (Vaughn et al., 2016). While a number of studies have estimated CH<sub>4</sub> fluxes from Arctic lakes and wetlands (Kodovska et al., 2016; Walter et al., 2006, 2008, 2013; Wik et al., 2013), few have focused on the Arctic marine shelves. One notable exception are estimates from the East Siberian Arctic Shelf, which indicated much higher fluxes (~8 Tg CH<sub>4</sub> y<sup>-1</sup>) emitted to the atmosphere than previously estimated for marine systems (Shakhova et al., 2010). These estimates suggest that atmospheric CH<sub>4</sub> fluxes from

*Abbreviations:* ACW, Alaska Coastal Water; AIC, Akaike information criterion; CH<sub>4</sub>, methane; COMIDA, Chukchi Sea Offshore Monitoring in Drilling Area; CTD, conductivity, temperature and depth; GC-FID, Gas Chromatography – Flame Ionization Detector; KOH, potassium hydroxide; OLS, ordinary least squares; SWI, sediment-water interface; TOC, total organic carbon; WC, water column

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<http://dx.doi.org/10.1016/j.dsr2.2017.01.004>

Available online 03 January 2017

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Arctic shelves could be significant and need to be quantified as part of the global budget.

While few studies have directly quantified CH<sub>4</sub> flux to the atmosphere from Arctic shelves, several studies have focused on overall CH<sub>4</sub> dynamics in these systems. For example, the earliest studies suggested that the Beaufort Sea, offshore of western Arctic Canada, could be a seasonal source of methane as the gas was present at highest concentrations under the ice (Kvenvolden et al., 1993; Macdonald, 1976). A more extensive survey of the area included measurements of methane in open water, sea ice, and bottom sediments (Lorenson and Kvenvolden, 1995). More recently, the same area was the focus of the MITAS (Methane in the Arctic Shelf/Slope) project to constrain sedimentary CH<sub>4</sub> fluxes associated with gas hydrate occurrence offshore in the Beaufort Sea (Coffin et al., 2013). They measured sedimentary diffusional CH<sub>4</sub> fluxes into the sulfate CH<sub>4</sub> transition zone ranging from 6 to 424 μmol CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>. These flux data, along with other geophysical information, indicated highly variable CH<sub>4</sub> fluxes within their sample area, which was consistent with an earlier suggestion that methane found in Beaufort Sea water originated from the sediments (Kvenvolden et al., 1993). Closer to the Bering Strait, water column microbial rates of CH<sub>4</sub> production and oxidation were measured in order to calculate an atmospheric flux of up to 57 μmol CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> (Savichev et al., 2007). This flux is approximately an order of magnitude less than that measured by Shakhova et al. (2010) on the East Siberian Sea Shelf.

Data constraining CH<sub>4</sub> fluxes are also available from around Svalbard, in both the Barents and Norwegian seas. In the Barents Sea (~350 m water depth), pockmarks, which are pitted scars left on the seafloor and could be indicators of an upward advective CH<sub>4</sub> flux, were studied to determine if they are currently charged with CH<sub>4</sub> (Nickel et al., 2012). While gas was present in the sediments, pore-water profiles of sulfate indicated little depletion with depth, suggesting very little flux of CH<sub>4</sub> to the sediment surface (Nickel et al., 2012). In deeper water (> 400 m), off the West Spitsbergen continental margin, gas flares emanate from the seafloor, possibly from decomposing gas hydrates (Westbrook et al., 2009). Surface waters above these flares however, contain low CH<sub>4</sub> concentrations (~20 nM) due to strong halocline stratification. This stratification is thought to enable aerobic microbial oxidizers to remove CH<sub>4</sub> before it diffuses into surface waters (Gentz et al., 2014). Thus, little CH<sub>4</sub> presumably reaches the atmosphere in Spitsbergen, as has been shown from other flare sites, especially those in deeper waters (McGinnis et al., 2006). A similar conclusion was drawn from the deep-water (1200 m) Håkon Mosby mud volcano but the authors suggest that CH<sub>4</sub> does not get oxidized in the water column, but instead is mixed and diluted (Damm and Budéus, 2003).

The studies summarized above indicate that the Arctic is a very heterogeneous region with a wide range of CH<sub>4</sub> concentrations and atmospheric fluxes from the shallow onshore and deeper offshore seafloor. While there is potential for a significant CH<sub>4</sub> flux from shallow shelf waters or even from ice-free polynyas in shallow waters (Damm et al., 2007), more sampling on a spatial and temporal basis is needed in order to fully understand current and potential future CH<sub>4</sub> emissions from the Arctic. Our goal with the present study is to fill this data gap and characterize dissolved CH<sub>4</sub> concentrations in the water column and surface sediments from a portion of the Chukchi Sea Shelf, which is spatially extensive, and considered to be one of the most biologically productive arctic shelves (Grebmeier et al., 2006). In addition, it has been estimated that about 75% of the offshore area of Alaska, including the Beaufort and Chukchi Seas, is underlain by gas hydrates, the largest reservoir of CH<sub>4</sub> on Earth (Collett, 2004; Gornitz and Fung, 1994). Thus, given the ongoing warming conditions, it is possible that significant or increasing CH<sub>4</sub> fluxes could originate from the Chukchi shelf sediments.

## 1.2. The Chukchi Sea shelf

The Chukchi Sea is relatively shallow (< 50 m) with two prominent shoals (Herald and Hanna Shoals) that are ~30 m in depth. Two water masses with Pacific water origins flow across the shelf from the Bering Strait (Fig. 1); a recent thorough review of the water masses is provided by Strong et al. (2016). Briefly, the first water mass is the Alaska Coastal Water, which advects low-salinity, low-nutrient, and low-organic carbon water to the eastern side of the sea including Barrow Canyon (Weingartner et al., 2005) and has a seasonally averaged primary productivity rate of approximately 50–80 g C m<sup>-2</sup> y<sup>-1</sup> (Grebmeier et al., 1995; Sambrotto et al., 1984; Walsh et al., 1989). The second water mass, Bering Sea water, moves to the west of the shelf and brings saltier, nutrient- and carbon-rich water with a primary productivity rate of 470–840 g C m<sup>-2</sup> y<sup>-1</sup> (Springer and McRoy, 1993). Bering Sea water varies seasonally, so that water passing through the Bering Strait in the summer open water period can be readily distinguished from water passing through the Strait during the ice covered period. This Bering Sea winter water, also termed Remnant Winter Water in the Strong et al. (2016) review, remains on the shelf in the summer, but has a lower temperature and higher salinity than water that has more recently passed through Bering Strait. The deep channel of Barrow Canyon on the northeastern portion of the Chukchi Sea is a convergence point for these waters as well as an entry point for the coastal water stratified above the more dense Bering Sea summer and winter waters as it reaches the shelf break. This Bering Sea water remains nutrient-rich as it traverses Barrow Canyon offshore and into the Arctic Ocean proper. Seasonal upwelling events in Barrow Canyon bring Arctic water with Atlantic characteristics (salinity > 33.1 psu) to the Chukchi shelf (Pickart et al., 2005 and references therein).

In the northeastern Chukchi Sea, currents flow clockwise around Hanna Shoal mixing Bering Sea Water from the west with dense winter water from the north of Hanna Shoal, Alaska Coastal Water from the south and freshened water bearing melted sea ice in our study area (Weingartner et al., 2013). These waters have a primary productivity rate of 80–90 g C m<sup>-2</sup> y<sup>-1</sup> (Grebmeier et al., 2006; Hill and Cota, 2005), and are typically stratified over Hanna Shoal during the open water period (Weingartner et al., 2005). For this study, we focused in the northeastern Chukchi Sea on Hanna Shoal and Barrow Canyon (Fig. 1), which is influenced by the Alaska Coastal Water as well as summer and winter Bering Sea water further offshore. Our work presented here was part of the Chukchi Sea Offshore Monitoring in Drilling Area (COMIDA)-Hanna Shoal Ecosystem study (Dunton et al., this volume), and also included sampling in Barrow Canyon as part of the international Distributed Biological Observatory (DBO) change detection array network on transect line DBO5 (Grebmeier et al., 2010, 2012). The COMIDA study overall was in part motivated by research needs to assess ecosystem status and biogeochemical processes prior to the initiation of oil and gas production on the Chukchi shelf. Although there are no longer any immediate plans for oil and gas production on the Chukchi shelf and exploratory drilling has been completed, the COMIDA studies are also meant to identify possible anthropogenic impacts from broader stresses such as climate changes associated with warming seawater and seasonal sea ice declines (Dunton et al., this volume).

## 2. Methods

### 2.1. Sample sites

Water and sediment samples were collected from the Hanna Shoal region and Barrow Canyon within the Chukchi Sea aboard the US Coast Guard Cutter *Healy* from 9 August to 25 August 2012 (Cruise name HLY 1201; Fig. 1). On Hanna Shoal, a general randomized tessellation stratified design was used to determine site locations (as described in

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