

Methane and nitrous oxide in surface water along the North-West Passage, Arctic Ocean

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ABSTRACT

Dissolved methane and nitrous oxide in seawater were measured along a 6700 km transect of the North-West Passage between the North Atlantic Ocean and Beaufort Sea in the Arctic Ocean. Over- and under-saturation with respect to atmospheric equilibrium were observed for both gases. Methane and nitrous oxide were in the range of 58–528% and 82–181% saturation, respectively. Under-saturation was attributed to melt-water with low methane and nitrous oxide, while over-saturation was found under multi-year sea-ice. Elevated methane was also found in the vicinity of the marginal ice zones and the Mackenzie River plume. Our data support both water column and sedimentary sources of methane and nitrous oxide. We found first-order methane oxidation in surface seawater with a rate constant of $3.8 \times 10^{-3} \text{ h}^{-1}$. Based on these results and a conceptual model, we suggest that future sea-ice retreat may decrease the residence times of methane and nitrous oxide in the surface Arctic Ocean and thus enhance the sea–air flux of these climatically active gases.

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1. Introduction

Methane (CH_4) and nitrous oxide (N_2O) are important biogenic trace gases with relatively long atmospheric lifetimes. They are infrared-active, together accounting for ~20% of enhanced greenhouse forcing (Forster et al., 2007). CH_4 is involved in the stratospheric water cycle and in the photochemistry of tropospheric hydroxyl radical (OH) and ozone (O_3) (Crutzen, 1991), and N_2O generates stratospheric NO_x which impacts O_3 (Nevison and Holland, 1997).

Constraining the marine contribution to tropospheric CH_4 and N_2O suffers from data scarcity in key areas and inherent uncertainties in scaling sea-to-air fluxes. Current best estimates are that marine waters provide <5% of tropospheric CH_4 and 29–55% of tropospheric N_2O ; the highest contributions are from estuaries and shallow coastal shelves (Bange et al., 1994, 1996; Bates et al., 1996; Seitzinger and Kroeze, 1998; Upstill-Goddard et al., 2000). Methanogenesis, an anaerobic microbial process mediated by archaea (Madigan and Martino, 2006), is thought to dominate marine CH_4 emissions, although cold seepage of thermogenic CH_4 and the dissolution of CH_4 hydrates may also be significant (Judd, 2004; Kvenvolden and Rogers, 2005). N_2O is a by-product of aerobic nitrification and an intermediate product of anaerobic denitrification (Hattori, 1983; Kaplan, 1983).

The Arctic Ocean is particularly sensitive to global warming through polar amplification and is thus recognised as one region where the resulting effects on ecosystems will be among the most dramatic (Denman et al., 2007; Holland and Bitz, 2003). Arctic sea ice thickness and extent are in decline due to increasing sea-surface temperatures (Comiso, 2002; Comiso et al., 2003; Gregory et al., 2002; Holland et al., 2006; Stroeve et al., 2007b; Teng et al., 2006; Wadhams, 1995). Sea ice presents a barrier to turbulent diffusion across the sea–air interface and hampers shelf-basin exchange by restricting the wave energy and thereby vertical diffusivity (Carmack and Chapman, 2003). Changes to the internal dynamics and sea–air fluxes of CH_4 and N_2O are therefore expected with decreasing sea ice cover. Additional CH_4 release might result from temperature destabilisation of gas hydrates on the shallow continental shelves (Westbrook et al., 2009), and organic matter stored in terrestrial and marine permafrost might be mobilised onto the shelves leading to further CH_4 and N_2O release through methanogenesis and denitrification.

As far as we are aware there is only one previously published N_2O dataset for the Arctic Ocean (Hirota et al., 2009), and existing CH_4 observations are restricted to localised studies that all showed strong CH_4 super-saturation relative to atmospheric air: the White Sea (Barents Sea) (Savvichev et al., 2004), the Beaufort Sea (Kvenvolden et al., 1993; Macdonald, 1976), the East Siberian Sea (Shakhova and Semiletov, 2007; Shakhova et al., 2005), and coastal waters off Spitsbergen (Damm et al., 2007, 2008).

In this paper we present data for CH_4 and N_2O collected during an approximately 7000 km transect from the south of Iceland, through

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Baffin Bay, the North-West Passage and the Beaufort Sea during July 2005. Due to the nature of the cruise being a transit leg for subsequent shore based fieldwork, sampling was necessarily limited to opportunistic surface underway (7 m) measurements. Nevertheless, we were able to also carry out a single 918 m hydrocast in Baffin Bay. Our measurements represent the first ever basin-wide survey of CH₄ and to the best of our knowledge only the second N₂O dataset from the Arctic Ocean.

2. Methods

Sampling was carried out onboard IB *Oden* during Beringia 2005 (4–28 July, 2005), a research initiative organized by the Swedish Polar Research Secretariat (<http://www.polar.se/beringia2005/eng/index.html>). The cruise track is shown in Fig. 1. Underway sampling for CH₄ and N₂O was in 1 L volumetric flasks with the exclusion of all bubbles, via a branch from the ship's pumped thermosalinograph supply (Sea-Bird, SBE 45; inlet 7 m). In order to remove ice-blockages, the supply's inlet was fitted with a back-flush system which was operated from the main lab following an audible alarm. Samples were only collected after the supply of seawater was restored for at least 5 min. Samples for dissolved silicate were similarly collected in polypropylene containers (NUNC cryotubes, cat. no. 379146) and stored at –20 °C until analysis by standard colorimetry (Hansen and Koroleff, 1999). In addition CH₄ and N₂O were sampled on a 918 m Niskin bottle hydrocast in Baffin Bay (73°44.78 N, 77°3.72 W) and at a surface melt-pond during an ice station (73°24.73 N, 96°18.13 W). Water depth at the hydrocast station was approximately 950 m.

Rates of microbial CH₄ and N₂O production/consumption were investigated by incubating surface seawater from under sea ice, and melt-pond water from an adjacent ice station. Incubations of headspace free duplicate samples were at 4 °C under fluorescent lighting, in gas-tight, 100 mL glass syringes (Samco), fitted with gas-tight, 3-way polycarbonate stopcocks. The melt-pond samples were collected directly from the pond edge in glass syringes while the seawater samples were collected with a 10 L Niskin bottle. On deck, the Niskin bottle was connected to a polycarbonate vessel fitted with a 0.45 µm filter at the outflow, thereby concentrating the plankton community in the vessel 15-fold by gravity filtration. Glass syringes containing the concentrated plankton community were then filled directly from the Niskin and incubated as above.

All water samples were analysed for CH₄ and N₂O by single phase equilibration (UHP N₂) gas chromatography with flame ionisation and electron capture detection for CH₄ and N₂O respectively; analytical precision for selected samples ($n = 3$) was better than ±2% for both

gases (Upstill-Goddard et al., 1996). For the incubation samples the procedure was modified to allow in-syringe equilibration (Upstill-Goddard et al., 1990). Method calibration used gravimetrically prepared primary standards (BOC gases Ltd.: 0.72 and 5.47 ppmv for CH₄ and 0.71 and 0.41 ppmv for N₂O). Each sample run was followed by a corresponding calibration standard and air sample collected from a gas line outside the bridge. Percent CH₄ and N₂O saturations relative to atmospheric air were calculated from the *in situ* concentrations and solubility data for CH₄ and N₂O (Weiss and Price, 1980). Some of our observations were below the temperature range of data on which these solubility functions were based (>–1 °C). Although this is not entirely satisfactory, there are currently no formulations for the required temperature range to our knowledge. Average mixing ratios of atmospheric CH₄ was 1.86 ppmv (range: 1.81–2.00 ppmv) and N₂O was 320 ppbv (range: 279–358 ppbv).

Daily sea-ice cover was obtained from the National Snow and Ice Data Centre (NSIDC; <http://www.nsidc.org>) Nimbus-7 SMMR and DMSP SSM/I 25 km resolution datasets (Knowles et al., 2002).

3. Results

An overview of the distribution of CH₄ and N₂O grouped into six geographic regions is given in Table 1. These regions were: a) the North Atlantic, b) Labrador Sea–Baffin Bay South, c) Baffin Bay Marginal Ice Zone, d) Baffin Bay North–Lancaster Sound, e) Canadian Archipelago and f) Amundsen Gulf–Beaufort Sea. The classification of the entire-dataset into these regions was based on geographic criteria as well as ice-cover characteristics. For example, Baffin Bay was broken down into three regions based on ice cover (see below and Fig. 2). Surface salinity showed an overall westward decrease along the transect (Fig. 2A). High variability to the west of Baffin Bay and into the Beaufort Sea reflects dilution by freshwater. A broadly inverse relationship between salinity and silicate (Fig. 2A and B) identifies this variability to result mostly from river discharge rather than localised ice melting; note for example the exceptionally low salinity and high silicate (>6.8 µmol L^{–1}) in the vicinity of the Mackenzie River Plume (MRP) 6200–6500 km along the transect (Figs. 1 and 2).

Dissolved CH₄ at this site exceeded atmospheric equilibrium (4.6 ± 1.0 nmol L^{–1}; $144 \pm 30\%$ saturation) and was higher than in the adjacent Beaufort Sea, consistent with the general observation of high CH₄ super-saturation in river outflow (Upstill-Goddard et al., 2000). Nevertheless, over the cruise track in general CH₄ did not appear to correlate closely with either salinity or silicate; evidently river discharge is not the major control on surface CH₄ concentrations throughout this

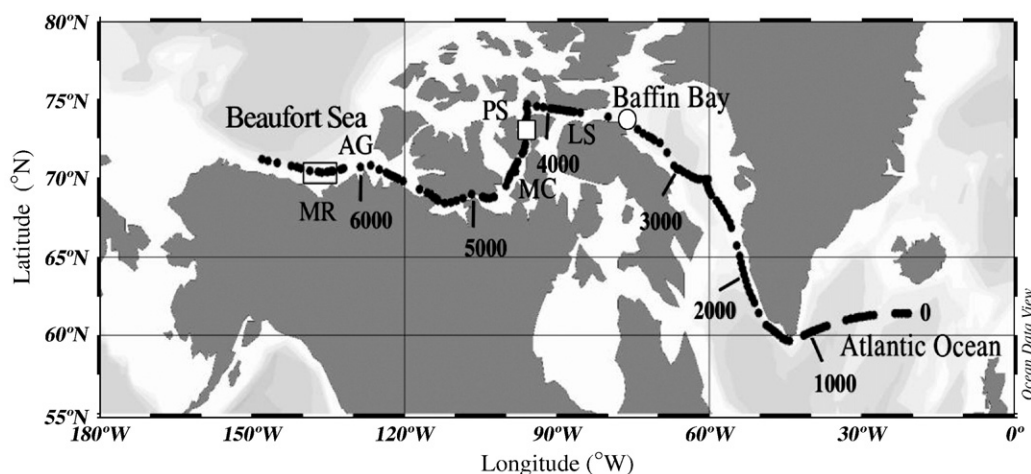


Fig. 1. Sampling locations (dots) along the cruise track of RV *Oden* with 1000 km markers from the first sample. The east-west cruise track crossed the North Atlantic Ocean, Baffin Bay, Lancaster Sound (LS), Peel Sound (PS), McClintock Channel (MC), Amundsen Gulf (AG), Mackenzie River plume (MR; in square) and Beaufort Sea. The figure was compiled using the Ocean Data View software. The locations of the Baffin Bay depth profile and ice station are marked by a white circle and square respectively.

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