Significant methane undersaturation during austral summer in the Ross Sea (Southern Ocean)

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Scientific Significance Statement

Oceans are considered to be a natural source of atmospheric methane (CH4). However, CH4 emissions in the high-latitude areas of the Southern Ocean are not yet well constrained. We found that the water in the Ross Sea is undersaturated with CH4 during austral summer, causing substantial CH4 uptake from the atmosphere, which highlights the unique role of high-latitude areas in assessing oceanic CH4 emissions.

Abstract

Methane (CH4) is a climate-relevant trace gas that is emitted from the open and coastal oceans in considerable amounts. However, its distribution in remote oceanic areas is largely unknown. To fill this knowledge gap, dissolved CH4 was measured at nine stations at 75°S in the Ross Sea during austral summer in January 2020. CH4 undersaturation (mean: 82 ± 20%) was found throughout the water column. In subsurface waters, the distribution of CH4 mainly resulted from mixing of water masses and in situ consumption, whereas the CH4 concentrations in the surface mixed layer were mainly driven by air-sea exchange and diapycnal diffusion between the surface and subsurface layers, as well as consumption of CH4. With a mean air-sea CH4 flux density of −0.44 ± 0.34 μmol m−2 d−1, the Ross Sea was a substantial sink for atmospheric CH4 during austral summer, which is in contrast with most oceanic regions, which are known sources.

Ice-covered parts of the Antarctic are now known to be a reservoir of organic carbon, which might fuel methane (CH4) production during organic matter degradation, suggesting that the Antarctic ice sheet may be a neglected but important component of the global CH4 budget (Wadham et al. 2012). However, the very limited number of observations of CH4 in the water column of the Southern Ocean constrains our ability to assess its oceanic CH4 emissions.

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The dynamics of CH$_4$ in the water column are driven by various processes. The exchange of CH$_4$ across the ocean–air interface is driven by the concentration gradient between both reservoirs; therefore, the CH$_4$ saturation in the surface mixed layer determines the direction of the CH$_4$ fluxes. In situ biological CH$_4$ production and consumption are major pathways that affect CH$_4$ concentrations in the water column (Reeburgh 2007). Sedimentary release of CH$_4$ from organic matter-rich sediments or by melting gas hydrates is another potential source of CH$_4$ in the water column. For example, evidence for the presence of gas hydrates in the western Ross Sea (Southern Ocean) was inferred from a bottom-simulating reflector, indicating a potential source of CH$_4$ under the seafloor (Geletti and Busetti 2011). Sea ice can be a physical obstruction for the air–sea exchange of CH$_4$, and the storage of CH$_4$ in ice crystals is usually a minor source compared to sea ice brines (bubbles or dissolved) and under-ice seawater (Crabeck et al. 2014; Zhou et al. 2014). Recently published first measurements of the CH$_4$ isotopic composition in Antarctic sea ice near Cape Evans revealed a potential combination of a hydrothermal source near Mount Erebus with a previously unknown CH$_4$ formation in sea ice (Jacques et al. 2021).

Only a few studies have addressed the distribution of CH$_4$ in the water column of the Southern Ocean (Lamontagne et al. 1974; Tilbrook and Karl 1999; Heeschen et al. 2004; Yoshida et al. 2011; Bui et al. 2018), and these studies have rarely focused on the Ross Sea. In the 1970s, surface CH$_4$ concentrations were investigated at several locations near the Ross Sea, showing a sink for atmospheric CH$_4$ that was explained by microbial consumption of CH$_4$ (Lamontagne et al. 1974). Hence, the CH$_4$-depleted regions of the Southern Ocean may act as CH$_4$ sinks, as reported for CO$_2$ in ice-free polar seas (Sandrin et al. 2007; Rysgaard et al. 2011). However, our knowledge about CH$_4$ cycling in the Ross Sea is rudimentary at best. To our knowledge, this is the first attempt to investigate the distribution of CH$_4$ in the Ross Sea since 1972 (Lamontagne et al. 1974). Findings from this study can offer new insights into CH$_4$ dynamics in high-latitude regions of the Southern Ocean.

**Methods**

**Study site and hydrographic measurements**

Seawater samples were collected onboard R/V “Xuelong 2” during the CHINARE 36$^{th}$ to the Ross Sea from 03 to 06 January 2020. The samples for the determination of dissolved CH$_4$ concentrations were collected at nine stations (R1–R7 and R9/R10) on a transect along 75°S from 164°E to 182°E (Fig. 1). The transect extends from Victoria Land to the middle of the Ross Sea shelf, covering several banks (water depth < 500 m) and troughs (water depth > 1000 m). Samples were taken from 8–10 depths at each station.

Hydrographic data were measured with an SBE-911$^{+}$plus conductivity–temperature–depth unit (Sea-Bird) that measured the salinity (± 0.002 psu), temperature (± 0.001°C), and pressure (0.015% of full-scale range). We classified five water masses (Text S1) and calculated the relative abundance of each water mass (Text S2) based on the hydrographic data. The Brunt–Väisälä frequency (cycles/h) was calculated following Pond and Pickard (1983). Monthly mean sea ice data in the Ross Sea and its adjacent areas were obtained from the Physical Sciences Laboratory, NOAA (http://www.psl.noaa.gov/data/gridded/data.ncep.reanalysis.derived.html).

**CH$_4$ sampling and analysis**

Seawater samples for CH$_4$ analysis (Ye 2022) were collected with a rosette water sampler equipped with Niskin bottles, following the sampling strategy described by Zhan et al. (2018) (Text S3). The CH$_4$ saturation (Sat, %) in seawater is calculated by the following equation:

$$Sat = \frac{C_{obs}}{C_{eq}} \times 100\%,$$

(1)

where $C_{obs}$ is the measured concentration of CH$_4$ in seawater, $C_{eq}$ is the concentration in equilibrium with the atmosphere calculated with the solubility equation of Wiesenburg and Guinasso (1979), in situ temperature (–1.9°C to 1.4°C) and salinity (33.68–34.84) at the time of sampling, and an atmospheric CH$_4$ dry mole fraction of 1.82 ppm (monthly mean during the sampling period at the South Pole Observatory;
Dlugokencky et al. 2020). The overall mean analytical error of $C_{\text{obs}}$ was ±0.3 nmol L$^{-1}$, with a corresponding mean error of ±10% for the CH$_4$ saturations.

Flux density calculations

The flux density ($F_{\text{ase}}$, μmol m$^{-2}$ d$^{-1}$) of CH$_4$ across the air–sea interface was estimated by the following equation:

$$F_{\text{ase}} = k_w \times (C_{\text{obs}} - C_{\text{eq}}),$$

(2)

where $k_w$ is the gas transfer coefficient (m d$^{-1}$) and $C_{\text{obs}}$ is the measurement from the ocean surface (2–5 m). We used an empirical model to determine $k_w$, which is a function of the molecular diffusivity of CH$_4$ in water (Jähne et al. 1987), the kinematic viscosity of water, and the wind speed at 10 m height (Wanninkhof 2014). Wind speeds (1.6–10.8 m s$^{-1}$) were measured by shipboard automatic weather stations, with a mean value of 5.4 ± 2.6 m s$^{-1}$. This value was in good agreement with the daily mean wind speeds (5.8 ± 0.1 m s$^{-1}$) over the Ross Sea during the sampling period obtained from the National Centers for Environmental Prediction (NCEP) reanalysis data set (Kalnay et al. 1996). Without considering the effect of wind speed, the uncertainties of $F_{\text{ase}}$ (±0.2 μmol m$^{-2}$ d$^{-1}$) were mainly caused by the error of $C_{\text{obs}}$.

CH$_4$ mass balance calculation in the surface mixed layer

The mixed layer was defined as the water depth where a potential density difference of 0.125 kg m$^{-3}$ and a temperature difference of 0.5°C in comparison with the ocean surface (<2 m) was computed (de Boyer Montégut et al. 2004). The CH$_4$ fluxes in the surface mixed layer should be balanced and can be expressed by the following equation:

$$-F_{\text{ase}} + F_{\text{vd}} + F_{\text{ad}} + F_x = 0,$$

(3)

where $F_{\text{ase}}$ is the air–sea exchange of CH$_4$ across the ocean/atm interface obtained from Eq. 2. $F_{\text{vd}}$ (μmol m$^{-2}$ d$^{-1}$) is the vertical (diapycnal) diffusion of CH$_4$, which can be obtained from Fick’s first law:

$$F_{\text{vd}} = K_z \times \frac{dc}{dz},$$

(4)

where $h$ is the depth (m), $dc/dz$ is the vertical CH$_4$ gradient measured at each station, and $K_z$ (m$^2$ s$^{-1}$) is the diapycnal diffusivity. We used a $K_z$ of 10$^{-4}$ m$^2$ s$^{-1}$, which was suggested to be representative of the Southern Ocean (Mashayek et al. 2017). $F_{\text{ad}}$ (μmol m$^{-2}$ d$^{-1}$) is the lateral CH$_4$ transport (advection) into/out of the surface mixed layer, which is influenced by both the external water intrusion and freshwater discharge from melting sea ice. Since only Antarctic surface water (AASW) was present in the surface mixed layers at all stations except of Sta. R1, we considered the influence of external water intrusion to be negligible. $F_x$ (μmol m$^{-2}$ d$^{-1}$) is the unknown in situ source/sink in the surface mixed layer, which was estimated by the balance of other flux terms.

Positive values of $F_x$ indicate a production of CH$_4$ in the surface mixed layer, while negative values indicate a consumption of CH$_4$ in the surface mixed layer. Eubullition may be a CH$_4$ source to the surface mixed layer (McGinnis et al. 2006), but especially in shallow shelf regions as reported by Shakhova et al. (2014) in the Arctic Ocean. However, the mass balance calculation did not consider the ebullition because of the lack of data to adequately address this process in our study.

Results

Hydrographic setting

Five water masses were identified (Fig. 2a). Shelf water (SW) was the dominant water mass in the study area west of 175°E, accounting for 60–90% of the total water volume (Fig. 2b; Tables S2, S3). In contrast, the contribution of modified shelf water (MSW) increased from ~5% in the west to ~20–50% east of the transect. Correspondingly, AASW and modified circumpolar deep water (MCDW) were found in the eastern part of the shelf, with the largest proportions of AASW and MCDW at Sta. R10 (65% for AASW and 34% for MCDW) and the lowest proportions (nearly zero) at Stas. R1 (for AASW) and R2 (for MCDW). Sta. R9 showed the most heterogeneous water mass composition, consisting of near equal contributions of AASW, MSW, and MCDW. A small fraction (10–19%) of ice shelf water (ISW) was found only close to the ice sheet (Stas. R1 and R3) and in the depth range between 200 and 300 m, where supercooled water could be formed in the cavities below the ice sheet.

CH$_4$ distribution along the transect

The CH$_4$ concentrations (saturation) in the water column along the transect were generally low (mean ± std: 3.1 ± 0.7 nmol L$^{-1}$) (82 ± 20%) and ranged from 1.5 to 5.5 nmol L$^{-1}$ (39 to 147%) (Table S6). Slightly higher mean CH$_4$ concentrations (saturation) (3.3 ± 0.7 nmol L$^{-1}$, 87 ± 19%) were found near the coast compared with those measured in the offshore areas (2.7 ± 0.7 nmol L$^{-1}$, 72 ± 19%, Stas. R6/R7 and R9/R10) (Fig. 3e,f). In the west, waters in the upper ocean had the highest CH$_4$ concentrations (saturation), with a mean of 3.5 ± 0.7 nmol L$^{-1}$ (94 ± 17%). Below 50 m, CH$_4$ concentrations (saturation) decreased to 3.3 ± 0.7 nmol L$^{-1}$ (84 ± 19%) at depths between 50 and 400 m and 3.2 ± 0.6 nmol L$^{-1}$ (84 ± 17%) below 400 m. In the east, Sta. R10 (where AASW dominated the water column) had the lowest CH$_4$ mean concentration (2.4 ± 0.3 nmol L$^{-1}$) and saturation (63 ± 8%) compared to the other stations. The highest CH$_4$ saturation (147%) was found at station R7 at 200 m, where anomalously warm water was also observed (0.7°C higher than ambient temperature).

CH$_4$ flux in the surface mixed layer

Two CH$_4$ mass balances were calculated for comparison (Fig. 4a): the western area (Stas. R1–R5) with more ice cover vs. the eastern area (Stas. R6–R10) with less ice cover (Fig. S3).
Fig. 2. Distribution of the main water masses in the Ross Sea during the austral summer of 2020. (a) Potential temperature $\theta$-salinity scatter plot for the sampling stations. Water masses are classified based on the temperature $\theta$-salinity definition (Table S1) as previously described (Orsi and Wiederwohl 2009; Schodlok et al. 2016; Williams et al. 2016; Zoccarato et al. 2016; Narayanan et al. 2019). Solid lines show the 28.00 and 28.25 kg m$^{-3}$ neutral density $\gamma_n$ surfaces (Jackett and Mcdougall 1997), which are used to define Antarctic surface water (AASW; lower bound) and to separate the circumpolar deep water (CDW) and modified circumpolar deep water (MCDW) from the Antarctic bottom water. The dashed horizontal line shows the surface freezing temperature of seawater, which separates the shelf water (SW) and ice shelf water (ISW). Dots are highly overlapping in the range of SW. (b) The proportions of water masses at each station.

Fig. 3. Vertical distributions of temperature (a), salinity (b), density (c), Brunt–Väisälä frequency (d), concentration (e), and CH$_4$ saturation (f) along the transect in the Ross Sea. The contour line of 100% CH$_4$ saturation (equilibrium with the atmosphere) is shown by the orange line. The potential source point of CH$_4$ released from sediment is marked with a red contour at Sta. R7 (see text). The white boxes indicate that no CH$_4$ data were available.
The Fad was positive in the west (0.37 μmol m⁻² d⁻¹) while it was negative in the east (0.27 μmol m⁻² d⁻¹). In the west, the air–sea exchange accounted for a mean uptake of 0.54 μmol m⁻² d⁻¹ from the atmosphere with a mean wind speed of 7.4 m s⁻¹. In contrast, the CH₄ uptake rate from the atmosphere was 0.32 μmol m⁻² d⁻¹ in the east, with a mean wind speed of 3.8 m s⁻¹. To simplify the calculations, we assume that the Fad at Sta. R3 was negligible because there was no external water intrusion (see CH₄ mass balance calculations in the surface mixed layer), and the approximately 90% sea ice concentration at Sta. R3 suggests negligible freshwater discharge from melting sea ice. Hence, the resulting rate of CH₄ oxidation (i.e., Fx) was 0.89 μmol m⁻² d⁻¹. A previous study reported a model-derived CH₄ oxidation rate for the Weddell Sea (Southern Ocean) ranging from 0.34 to 1.03 μmol m⁻² d⁻¹ (Heeschen et al. 2004), which is in good agreement with our result. Consequently, the mean Fad can be calculated from the mass balance (Table S6), which yields mean values of −0.02 μmol m⁻² d⁻¹ in the west and 0.84 μmol m⁻² d⁻¹ in the east.

Discussion

Role of water mass mixing in the CH₄ distribution

CH₄ was generally undersaturated or close to the equilibrium concentration, except for the bottom layer at R7. Our data are, therefore, in good agreement with published CH₄ measurements from the Southern Ocean (Table 1) and underline the fact that the waters of the Southern Ocean are generally undersaturated with CH₄. The unusually enhanced CH₄ concentration in the bottom layer at R7 indicates the presence of other CH₄ sources on the seafloor, and we suggest that the sediment may be a potential explanation. The substantial CH₄ undersaturation observed during our study in the Ross Sea indicates, on the one hand, that there was no appreciable in situ production of CH₄ and, on the other hand, that the CH₄ distribution mainly resulted from a combination of water mass mixing, air–sea exchange and microbial consumption. A mixing diagram (Fig. S2) showed that the CH₄ concentrations were lowest in the deep waters (e.g., circumpolar deep water [CDW] or Weddell Sea deep water [WSDW]) (Heeschen et al. 2004) at the edge of the Ross Sea. We found that the relatively lower CH₄ concentrations at the east part of the transect (compared to the west part) corresponded with the higher percentages of AASW and MCDW (Figs. 2b, 3e), indicating the impact of MCDW and AASW on the CH₄ distribution. High Brunt–Väisälä frequencies (> 15 cycles/h) were found near the coast (Stas. R1–R5) in the upper ocean (0–50 m), while the values were more irregular at offshore stations R6–R10 (Fig. 3d). In view of the pronounced water column stratification and a relatively slow water mass turnover time of 4 yr residence time for SW (Trumbore et al. 1991), we assumed that the mean CH₄ concentration of 3.2 nmol L⁻¹ in the deep waters below 400 m represents the bulk CH₄ deep water concentration in early summer. Consequently, we found that mixing AASW, SW, and CDW (substituted by WSDW in Fig. S2) could decrease the bulk CH₄ deep water concentration by 13–53% and thus lead to a higher CH₄ undersaturation in the Ross Sea. It should be
Table 1. Summary of saturation and air-sea flux densities of CH4 in the Southern Ocean (60–75°S).

| Study area                  | Date           | Latitude | Atmospheric mol fraction, ppm | Saturation, % | Flux density, μmol m⁻² d⁻¹ | References                  |
|-----------------------------|----------------|----------|------------------------------|---------------|----------------------------|------------------------------|
| Ross Sea                    | Dec 1972       | 70–77°S  | 1.36                         | 64            | −1.1*                      | Lamontagne et al. (1974)     |
| Antarctic peninsula (offshore) | Dec 1986–Mar 1987 | 62–64°S | 1.51                         | 87            | −0.35                      | Tilbrook and Karl (1994)     |
| Weddell Sea                 | Mar–May 1998   | 60°S     | 1.69                         | 75–94         | −0.5                       | Heeschen et al. (2004)       |
| Adélie Coast                | Dec 2001–Feb 2002 | 60–66°S | 1.71                         | 87–130        | −1.0 to 1.2*                | Yoshida et al. (2011)        |
| Indian/Pacific Ocean sectors | Dec 2012–Feb 2013 | 60–65°S | 1.75                         | 88–105        | −3.9 to 0.5                 | Bui et al. (2018)            |
| Ross Sea                    | Jan 2020       | 75°S     | 1.82                         | 86            | −0.44                      | This study                   |

*Values computed based on data provided in the corresponding study. The atmospheric mixing ratio and NCEP reanalysis wind speed taken from the sampling year were used in the flux estimation.

noted that the microbial oxidation of CH4 is a prerequisite for CH4 undersaturation in deep water and causes CH4-depleted surface waters (Tilbrook and Karl 1994; Heeschen et al. 2004), when water stratifications is eroded and vertical mixing occurs. Hence, superimposed on the effect of microbial oxidation of CH4, mixing between different water masses determines the distribution of CH4 in the Ross Sea.

CH4 dynamics in the surface mixed layer

The surface mixed layer plays an important role in connecting the atmosphere and the deep ocean. The air-sea exchange was comparable among the various stations. However, the vertical diffusion of CH4 was a source (upward flux) to the mixed layer in the west but changed to a sink (downward flux) in the east (Fig. 4a). In the west, uptake by air-sea exchange accounted for approximately 59% of CH4, while CH4 oxidation was the main process (98%) involved in the removal of CH4 from the surface mixed layer. In the east, while CH4 oxidation was still the main process (76%) involved in maintaining the status of surface CH4 undersaturation, the advection of CH4 accounted for 72% of the CH4 supply to the surface mixed layer.

Sea ice melt may be the dominant factor that influences Fad in the surface mixed layer. Fad will be a source due to the release of brine-enriched CH4 from sea ice (Damm et al. 2015) while becoming a sink due to the dilution of depleted CH4 from freshwater. In our cases, we found that Fad was a minor sink in the west and a source in the east, which resulted from the counterbalance between the effect of CH4 release and freshwater dilution. Importantly, the calculated Fad was negatively correlated with the ice concentration (r² = 0.67, p < 0.05; Fig. 4b), which implies that the sea ice may be a source of dissolved CH4 during the early stage of sea ice melt. An explanation is that sea ice might act as a partial barrier for gas exchange, and thus, CH4 accumulates in below-ice seawater during the ice-covered period (winter) (Damm et al. 2007). When sea ice basal melt begins, the permeability of the ice surface is still low, dampening gas exchange. This in turn might result in CH4 release into under-ice seawater (Damm et al. 2015). During the latest stage of sea ice melt, dilution may take place because sea ice is depleted in brine and CH4. It should be noted that the contribution of sea ice melt to the CH4 concentration may be minor if there is a deep surface mixed layer depth, for example, 29–39 m at Stas. R9–R10 (Table S3), or the dissolved CH4 is dominated by strong biological processes, for example, CH4 oxidation. In addition, gas exchange would be restricted even in partially ice-covered regions (Rutgers van der Loeff et al. 2014). Hence, sea ice freezing and melting cycles are important factors that affect CH4 dynamics in the surface mixed layer, but microbial consumption of CH4 is likely the key factor that determines how much CH4 could be taken up from the atmosphere during ice-free periods.

CH4 uptake in the Southern Ocean

Unlike most oceanic areas, where supersaturation of CH4 is found in the surface water (Dean et al. 2018), the Ross Sea was undersaturated at the time of sampling (austral summer) and was therefore characterized by an uptake of CH4 from the atmosphere. Unfortunately, to date, there are no CH4 data from fall and winter in the Ross Sea; therefore, by using the summer data, we calculated a mean CH4 flux density of −0.44 ± 0.34 μmol m⁻² d⁻¹ (with a range from −1.26 to −0.22 μmol m⁻² d⁻¹) and a CH4 uptake of 0.0006 Tg (extrapolated to a surface area of 9.6 × 10⁵ km²), which indicates that the Ross Sea was, similar to other regions from the Southern Ocean, a sink for atmospheric CH4 during ice-free periods (Table 1). Combing results from Table 1, a total of 0.02 Tg CH4 was calculated to be taken up during the three ice-free months (December–February) by the Southern Ocean surface seawater, accounting for 0.2–0.3% of the global oceanic CH4.
release (Weber et al. 2019). However, no CH4 data from other seasons are available at present, which hampers the calculation of an annual CH4 emission estimate.

Despite the increase in the dry mole fractions of atmospheric CH4 in recent decades (Table 1), our results indeed show that the high-latitude regions of the Southern Ocean still maintain the potential to take up atmospheric CH4 during austral summer. We anticipate that studies conducted during both summer and winter will lead to an improved understanding of the underlying mechanisms that determine the water column distribution of CH4 as well as the magnitude and variability of CH4 uptake in the Southern Ocean. These insights may be important for evaluating the role of the Southern Ocean in local or global CH4 budgets.

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