Methane release from open leads and new ice following an Arctic winter storm event

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Abstract

We examine an Arctic winter storm event, which led to ice break–up, the formation of open leads, and the subsequent freezing of these leads. The methane (CH_4) concentration in under–ice surface water before and during the storm event was 8–12 nmol L^{-1}, which resulted in a potential sea–to–air CH_4 flux ranging from +0.2 to +2.1 mg CH_4 m^{-2} d^{-1} in open leads. CH_4 ventilation between seawater and atmosphere occurred when both open water fraction and wind speed increased. Over the nine days after the storm, sea ice grew 27 cm thick. Initially, CH_4 concentrations in the sea ice brine were above the equilibrium with the atmosphere. As the ice grew thicker, most of the CH_4 was lost from upper layers of sea ice into the atmosphere, implying continued CH_4 evasion after the leads were ice–covered. This suggests that wintertime CH_4 emissions need to be better constrained.

1. Introduction

CH_4 emissions in a warming Arctic climate are suggested to increase gradually (Shuur et al., 2015). Arctic Ocean (AO) waters, which are largely covered by sea ice, receive CH_4 gas from numerous geological sources, such as dissociating gas hydrates (Paull et al., 2007; Westbrook et al., 2009), gas reservoirs (e.g., sub-sea and land-based hydrocarbon seeps) (Portnov et al., 2016; Platt et al., 2018), and decaying submarine permafrost (Portnov et al., 2013). Depending on the strength of geological plume and water depth, some of the CH_4 released at seafloor reaches the surface waters (Shakhova et al., 2010; Graves et al., 2017; Silyakova et al., 2020; Thornton et al., 2020). Surface AO waters are also known for biologically produced CH_4 excess due to nutrient limitation (Damm et al., 2010; 2015a) and because of physical rejection of brine containing CH_4 during sea ice growth and downward brine flushing with meltwater during sea ice melt (Damm et al., 2015b).

Different inputs result in an increase of CH_4 concentration in the water column. Increasing CH_4 concentration from deep to surface waters and surface water CH_4 super–saturation was found in ice–covered regions of the AO (e.g., Kvenvolden et al., 2003; Fenwick et al., 2017). While surface stratification hinders excess CH_4 from mixing with deeper waters (Damm et al., 2015b), sea ice, on the other hand, hampers direct and rapid CH_4 release to the atmosphere. Therefore, CH_4 accumulates and resides in under–ice surface waters for prolonged periods (Kitidis et al., 2010). As a result, a greater concentration of dissolved CH_4 in waters below the sea ice cover when compared to open waters is found in many regions of the AO (e.g., Kvenvolden et al., 1993; Kitidis et al., 2010; Shakhova et al., 2010; Shakhova et al., 2015; Damm et al., 2015b).

CH_4 accumulates beneath sea ice in the AO waters (Damm et al., 2018) before being oxidized by microbes (Damm et al., 2007; Kitidis et al., 2010; Damm et al., 2015a), which is the primary removal mechanism of CH_4 in ocean waters (Reeburgh, 2007), or released into the atmosphere through fractures in sea ice. The latter could potentially be a significant CH_4 source to the atmosphere in the Arctic (Kort et al., 2012). Climate change increases the mean speed and deformation of the Arctic sea ice, which results in an increasing amount of fractures in the ice pack (Rampal et al., 2009). Arctic storms contribute to fractures of summer multi–year sea ice (Asplin et al., 2014), and fracturing can also increase in the thinner and younger Arctic ice pack in winter (Itkin et al., 2017), which in turn increases the potential for winter air–sea gas exchange (Fransson et al., 2017). Further high wind speeds during storms promote the gas exchange processes at the air–sea interface in open water leads (Wanninkhof, 2014).

In this paper, we focus on CH_4 dynamics in under–ice water and new thin sea ice formed in open water leads during and after a major winter storm in February 2015 in the Nansen Basin of the AO. We report concentrations of dissolved CH_4 in under–ice surface water and post–storm formed thin sea ice; CH_4 temporal dynamics within sea ice over six days; and estimated the sea–to–air CH_4 flux from open water leads at the time of ice break up during the storm. This implies that wintertime fracturing of the otherwise rather impermeable ice pack can result in significant CH_4 fluxes that need to be better constrained to understand their role in the Arctic CH_4 budget.

2. Methods

2.1. N–ICE2015 campaign and the major storm event

The Norwegian research vessel (R/V) Lance froze into the Arctic ice pack in the Arctic Ocean's Atlantic sector in January 2015 to study the environmental processes until June same year as part of the Norwegian young ICE (N–ICE2015) campaign. The data used in this study was collected when R/V Lance was anchored to Floe 1 of N–ICE2015 from January 15 to February 21 when

newly formed thin sea ice covered open leads after the major storm event (Fig. 1). The major N–ICE2015 storm event M2 started on February 3 and ended on February 8 (Cohen et al., 2017). This storm was specified as a winter storm (Itkin et al., 2018). Atmospheric pressure decreased by 14 hPa in 6 hours, peak wind speed at 10 m height was 22 m s^{-1}, and the air temperature increased from –35.5°C to –1.4°C in the early phase of the storm but dropped quickly down to –30°C (Fig. 2). Conditions led to ice break up and formation of multiple open water leads (Fig. 3A, B) with their subsequent freezing.

Ice coring, seawater sampling, sea ice and snow observations (Rösel et al., 2018), and meteorological observations (Cohen et al., 2017) were accomplished from an ice camp situated on the ice floe 300–400 meters away from the R/V Lance. Granskog et al. (2016; 2018) present a detailed description and motivation of the campaign.

2.2. Under–ice water sampling

Under–ice water was sampled using a Hydro–Bios SlimLine 6 CTD equipped with an integrated CT–set and six 3.5 L sample bottles before and after the storm on February 2, 3, 9, and 10. Sampling took place in a tent 400 meters away from the ship. Seawater was collected from different depths from the surface down to 1000 m. Seawater for dissolved CH_4 analysis was transferred from the sample bottle into a 160 mL serum bottle using silicon tubing. Before filling, the bottles were rinsed three times with sample water, avoiding air bubbles. The sample was then poisoned with 50 µL of saturated $HgCl_2$, closed with isobutyl septa, crimped, and stored at +4°C and dark until analysis. Under–ice water CH_4 concentrations are only reported for the upper 70 m below the sea ice.

2.3. Ice coring and sampling

New ice was sampled in two refrozen leads, and both are representative of the early ice formation (see Fig. 3 for the location of the two leads). Lead sampled on February 7 was a few tens meters wide. This floe area experienced divergent motion between February 2 and 7, and multiple fractures opened, closed, compressed, and sheared. A larger lead was sampled between February 9 and 12 (Fig. 3B). Six ice cores were collected on February 7, 9, and 10, and three on 12 (Table 1), given the homogeneous characteristics if this new ice, we believe this sampling provides appropriate representation of the ice cover in the leads over time. Ice cores on a single day were sampled 10–20 m apart along the edge of the lead. When the ice was thinner than 20 cm on February 7 and 9, a saw was used to cut out large squares of thin ice, which were temporarily placed in a bucket with an air–tight lid on for transportation to the ship and further processing. Before sampling, the ice temperature (T) was measured at the ice surface and in the middle of the ice using an electric drill and a calibrated probe (Testo 110 NTC, Brandt Instruments, Inc., USA). There were no ice T measurements on February 7. Therefore, assuming linear T gradient across thin ice, we estimated the ice T on February 7 based on the T gradient measured on February 9 with bottom ice T of –1.9 °C, a freezing point at a salinity of 34.3 in under–ice surface waters on February 9.

For ice thicker than 20 cm (February 10 and 12), ice cores were collected using Kovacs ice corer with an internal diameter of either 9 or 14 cm (Mark II or Mark V, Kovacs Ent., Rosenburg, USA). Ice T was measured at every 10 cm of the core immediately after recovery. Each core for CH_4 measurements was cut into 10 cm sections and temporarily packed into Ziplock® bags for immediate transfer to R/V Lance. Onboard, all ice sections and pieces were immediately transferred into gas-tight Tedlar® plastic bags (5L Smart bag, GL Science, Japan), vacuumed by

using a syringe, and left to melt at +4°C in the dark. For CH_4 measurements, melted water from a gas–tight bag was transferred into a 60 ml serum bottle, using a silicon tube, poisoned with 50 µL of saturated $HgCl_2$, closed with isobutyl septa, crimped, and stored at +4°C until analysis. For salinity measurements, melted water was put into glass bottles and measured onboard using a salinometer (Guildline 8410A, Canada) with an accuracy of ca. ±0.003.

2.4. Methane analysis

CH_4 concentrations in melted sea ice section (bulk ice CH_4 concentrations) and seawater samples were determined using the headspace technique within a few months after sampling (Upstill–Goddard et al., 1996). Headspace in melted sea ice samples, was analysed by a gas chromatograph (GC, SRI® 8610) equipped with a Flame Ionization Detector (FID). For gas chromatographic separation, we used a packed column (Hayesep D). The GC oven was operated isothermally (+50°C), and the FID was held at +340°C. After creating a 25 mL N_2 headspace in 60 mL glass serum bottles and 30 mL in 160 mL bottles, samples were vigorously shaken for 20 minutes and placed in a thermostatic bath overnight at –1.6°C. The following day, the samples were shaken again for 20 minutes before the GC analysis. CH_4:CO_2:N_2O mixtures in the N_2 balance gas (Air Liquide, Belgium) of 1, 10, and 30 ppm of CH_4 were used to create a three–point calibration curve for a standard. For under–ice seawater samples from February 2, 3, 9, and 10, we used gas chromatograph Agilent GC7890A with a FID. After creating 5 mL N_2 headspace in 160 mL serum bottles, samples were vigorously shaken on a shaker while brought to lab temperature (+20°C). For gas chromatographic separation, we used a packed column (Porapac Q 80/100 mesh). The GC oven was operated isothermally (+60°C), and the FID was held at +200°C. Two sets of standard gas mixtures were used for calibration. The standard deviation of duplicate analyses was 5%. This overall error is almost exclusively due to the gas extraction procedure. The accuracy of the measurements by both instruments was within 3%. Final concentrations were computed using the CH_4 solubility coefficients given by Wiesenburg and Guinassio (1979).

2.5. CH_4 saturation and brine volume

CH_4 saturation in seawater, $CH_{4\,sat}$ (%) was computed following equation 1:

$$CH_{4\,sat} = (C_m/C^*) \cdot 100, \quad \text{(Eq. 1)}$$

where C_m is measured CH_4 concentration in seawater or sea ice, and C* is calculated CH_4 concentration at equilibrium with the atmosphere (4 nmol L^{-1}, with salinity, S = 34.3 and seawater temperature Tsw = –1.88°C and atmospheric CH_4 mole fraction of 1900 ppb, Zeppelin Observatory, Svalbard on February 3, 2015) following Wiesenburg and Guinasso (1979). If $CH_{4\,sat}$ is higher than 100%, seawater or sea ice are CH_4 super–saturation and above equilibrium concentration with the atmosphere.

Brine volume fraction, as function of salinity and temperature, was calculated using in situ temperature and bulk salinity following Cox and Weeks (1973). Sea ice permeability is defined by a brine volume threshold of 5% (Golden et al., 1998). If brine volume is less than 5%, sea ice is impermeable.

2.6. Sea–to–air CH_4 flux calculations

The sea–to–air CH_4 flux F was calculated according to Wanninkhof et al. (2009):

$$F = k_{660} \cdot (C_m - C^*), \text{ (Eq. 2)}$$

where k_{660} is the calculated gas transfer velocity (cm hr^{-1} Table 2, Fig. 4; or m d^{-1} used in Eq. 2), C*, and C_m are in nmol m^{-3}. k_{660} is normalized to a Schmidt number of 660, which is the ratio of water viscosity to molecular diffusivity.

For flux calculations in this study, we compare different parametrizations of gas transfer velocity k_{660} (Table 2 and Fig. 4), including scaling of k_{660} from Wanninkhof (2014) and Butterworth and Miller (2016) to the fraction of open water (f), which resulted in $k_{eff} = k_{660} \cdot f$ as suggested by Loose et al. (2014, 2016) and as in Butterworth and Miller (2016) for sea–ice zones. In Fig. 4, u indicated the measured wind speed at the meteorological mast at 10 m above the sea ice surface, which ranged between 0 and 25 m s^{-1}. Component f was calculated as the sea–ice fraction area subtracted from one. The sea–ice fraction was obtained from the AMSR2 microwave radiometer on the JAXA GCOM–W satellite. Sea–ice concentrations were derived from the 89 GHz channels, which allow a daily full global coverage of all sea–ice areas on a 6.25 x 6.25 km^2 grid (Spreen et al., 2008). The mean sea–ice concentration for a square of 43.75 x 43.75 km^2 (7 x 7 grid cells) with R/V Lance in the center pixel was calculated on an hourly basis. The GPS position of R/V Lance was used to identify the center grid cell in the ice concentration data set.

Since there was no significant difference between W14 and B&M16 (Fig. 4), we present fluxes calculated with k_{660} parametrization for open water based on (1) Wanninkhof (2014), (W14), (2) k_{660} parametrization for mixed sea ice/open water (lead site) from Prytherch (2020), (Pr20), and (3) Wanninkhof (2014) taking into account open water fraction according to $k_{eff} = k_{660} \cdot f$ (W14 Owfr).

3. Results and Discussion

3.1. CH_4 super–saturation in under–ice water

Observed under–ice water CH_4 concentration of 8–12 nmol L^{-1} equals $CH_{4\,sat}$ of 200–300%, which is super–saturation and above equilibrium concentration with the atmosphere (4 nmol L^{-1}, see Methods 2.5) (Fig. 5). This super–saturation is in agreement with observations of under–ice water below drifting sea ice from other AO regions (e.g., Kitidis et al., 2010; Damm et al. 2015a; Fenwick et al., 2017; Verdugo et al., 2021) and can potentially drive significant sea–to–air fluxes of CH_4.

3.2. Physico–chemical characteristics and methane evolution in newly formed sea ice

Newly formed sea ice rapidly covered open water leads following air temperature drop from –2°C to –33°C (Fig. 2). Once formed, this ice did not break as the meteorological conditions were stable (Fig. 2). The thickness of newly formed sea ice in the lead on February 7 was 8 cm, and in another lead increased from 18 to 27 cm in 4 days from February 9 to 12 (Fig. 6 and 7).

On the first day of observation (February 2), newly formed sea ice had high salinity (15–18, Fig. 6A), high temperature for the top (–7.2°C, Fig. 6B), and high brine volume for top (10–13%, Fig.

6C) with respect to those on the subsequent days. This is typical for young ice forming over open leads in winter (Perovich and Gow, 1996). This ice was very porous, which allows gas exchange through the growing ice since it highly permeable. Gases dissolved in brine are rejected downward to under–ice water together with brine during ice formation (e.g., Weeks and Ackley, 1986; Vancoppenolle et al., 2013) but at same rate as salts. This is implied by the lower sea ice salinity at the bottom ice section on February 12 (salinity 10–12), which grew later when compared to top horizons (salinity 13–17) (Fig. 6A). Higher CH_4 concentrations in bottom ice sections compared to top horizons (Fig. 7A) suggest the downward movement of CH_4 containing brine (Damm et al., 2015b). On the other hand, higher CH_4 concentrations in the bottom ice sections could also be explained by the fact that new ice grown underneath contains a high amount of CH_4 from supersaturated under–ice seawater. However, diffusive gas flux from under–ice water into the sea ice (across the concentration gradient between "brine" in bottom of sea ice and under-ice water) was shown to be negligible (2%, Lovely et al., 2015) and hence is unlikely to contribute to higher CH_4 concentrations in bottom sections of growing sea ice.

The top of the ice was covered with frost flowers (Fig. 3C), which is a sign of brine being expelled upwards to the ice surface (e.g., Perovich and Richter-Menge, 1994; Barber et al., 2014) and gases leaving sea ice into the atmosphere (Fransson et al., 2015, Granfors et al., 2015, Nomura et al. 2018). Higher salinity in the top of the ice compared to bottom horizons was observed in this study (Fig. 6A), even in later days of observations, could indicate upward ejection of brine (Kaleschke et al., 2004). At the same time, the CH_4 concentration decreases (from 5–7 nmol L^{-1} to 4 nmol L^{-1} over five days (Fig. 7A) and CH_4 to salinity ratio decrease in top horizons of newly formed sea ice (Fig. 7B) suggests that upper layers lost CH_4 into the atmosphere relative to salts. CH_4 release to the atmosphere occurs due to the diffusion of dissolved gas through the equilibration between the brine in the top of the ice and the atmosphere without exchange of salt. Also, CH_4 containing buoyant bubbles that are trapped in seawater during sea–ice formation travel upward in the ice through brine channels and release to the atmosphere (e.g., Loose et al., 2009; 2011; Crabeck et al., 2014a). Moreover, bubbles are formed within sea ice structure when CH_4 solubility lowers due to the increase of salinity in brines (Zhou et al., 2013, Zhou et al., 2014). Bubble formation is likely to be enhanced in young ice as there is still a large volume of concentrated salty brine that lowers solubility (Zhou et al., 2013), and the ice is permeable. As brine volumes stayed significantly higher than the 5% permeability threshold in the upper layer of the ice for all sampling days (Fig. 6C), there was a potential for continuous CH_4 evasion from the brine in the upper ice horizons. Based on our observations, we surmise that the thin ice formed after the winter storm was porous and an active source of CH_4 into the atmosphere. This finding agrees with elevated CO_2 fluxes from thin ice observed by Nomura et al. (2018).

3.3. Sea–to–air CH_4 flux in open leads

Open water leads frequently appeared between the beginning of the storm on February 3 and the last day of ice coring in this study on February 12, as indicated from radar images (Haapala et al., 2017). During the storm, calculated mean sea–to–air CH_4 flux from these open water leads was +0.31 mg CH_4 m^{-2} d^{-1} with a maximum flux of +1.59 mg CH_4 m^{-2} d^{-1} with a surface water CH_4 concentration of 10 nmol L^{-1} (based on open water parametrization of k_{660} W14, Fig. 5 top, Table 3) (Fig. 8). For the calmer post–storm conditions, the mean CH_4 flux was +0.08 mg CH_4 m^{-2} d^{-1}, and the maximum flux was +0.13 mg CH_4 m^{-2} d^{-1}. Thus, the highest flux was estimated during the storm at high wind speeds. After the storm, in calmer weather, the flux from open leads decreased as wind speeds decreased, and the leads froze over (Fig. 8).

In the open ocean, where the difference between surface water and atmospheric CH_4 concentrations is not very large, the flux depends mainly on wind speed, since the deciding part of the equation, the gas transfer velocity k_{660}, depends on wind speed. Comparing five different k_{660} parametrizations for the open ocean, Graves et al. (2015) concluded that different k_{660} parametrizations yield overall sea–to–air CH_4 fluxes ranging from 20 to 35% lower and 30 to 75% higher than mean flux, depending on the wind speed.

Flux calculations in the open leads show the same as in the open ocean dependency on wind speed. Prytherch and Yelland (2021) proposed that gas transfer in sea ice–covered areas mixed with open water leads is decreased by 25% relative to the open ocean (based on eddy covariance measurements of CO_2 fluxes in the central AO). Using Pr20, we calculated mean and maximum CH_4 fluxes during the storm as 0.23 and 1.20 mg CH_4 m^{-2} d^{-1}, respectively, while in calm weather as 0.06 and 0.1 mg CH_4 m^{-2} d^{-1} respectively (Fig. 8).

In the presence of sea ice, Loose et al. (2014, 2016) suggested that air–sea gas exchange not only depends on wind speed but on sea–ice fraction itself, surface water currents, and convection–driven turbulent mixing. The latter two are suggested to drive the air–sea gas exchange in the way of replenishing surface waters supplying excess gas to surface water open to the air, thus more gas to be released into the atmosphere (Damm et al., 2007; Lovely et al., 2015; Damm et al., 2015a; Loose et al., 2016). Prytherch and Yelland (2021) observed, however, that this convection–driven turbulent mixing is less likely to influence gas exchange in the sea ice–covered areas with open leads in the central AO in late summer.

Following the approach of scaling CH_4 flux to the open water fraction (Loose et al., 2014) implies that CH_4 transfer only occurs in the open water leads. During the storm event in this study, the open water fraction around R/V Lance in an area of 43.75 km^2 increased from 5 to 30% (Fig. 4). Fluxes scaled to the open water fraction (W14 OWfr) were 91 and 87% lower than fluxes based on open water parametrization W14 and sea ice/open leads parametrization Pr20, respectively (Table 3) because scaling CH_4 flux to the open water fraction (W14 OWfr) does not take into account the CH_4 exchange for the sea ice area, and the presence of sea ice reduces the gas exchange process. Therefore, Pr20 parametrization is valid for our study area, which also had a sea ice cover with open water leads.

Scaling CH_4 flux to the open water fraction implies that no CH_4 exchange occurs through sea ice (Kitidis et al., 2010). Despite the upward diffusion of gas from under–ice water to sea ice might be negligible (Lovely et al., 2015), direct measurements of CO_2 fluxes on sea ice suggested that gas exchange through the brine channels within sea ice is significant (e.g., Delille et al., 2014; Nomura et al., 2018). However, similar direct measurements for CH_4 fluxes are few. He et al. (2013) (summer in central AO, –0.94 to +0.77 mg CH_4 m^{-2} d^{-1}) and Nomura et al. (2020; 2022) (Lake Saroma, +0.01 mg CH_4 m^{-2} d^{-1}) measured CH_4 fluxes from sea ice to the atmosphere with the chamber technique. Remarkably, measurements in the central AO indicate not only positive but also negative CH_4 flux, implying that sea ice is not always a source but can also be a sink for atmospheric CH_4 since sea ice has lost CH_4 to the atmosphere (and partly ocean below), it can become a potential sink. In addition, especially summer-time, snow/sea ice meltwater dilute the CH_4 at the surface of sea ice and decreases CH_4 concentration with respect to the atmosphere. Therefore, sea ice could act as a potential sink for atmospheric CH_4. This CH_4 seasonal variation agrees with that of CO_2 concentration within the sea ice and flux between sea ice and atmosphere (e.g., Delille et al., 2014).

Despite the evidence of CH_4 exchange across the surface of sea ice, most studies reporting marine CH_4 fluxes in the AO are based on k_{660} parametrizations for the open ocean in the ice–free zones and assume no CH_4 flux through the sea ice cover (Table 4). Moreover, it appears that the CH_4

flux is higher in AO regions with CH_4 supersaturated surface waters (Thornton et al., 2016) connected to a geological sources. Areas with degrading subsea permafrost as the Laptev, East Siberian, and Chukchi Seas emit the most CH_4 to the atmosphere in ice–free conditions (on average 1.5 to 3.8 mg CH_4 m^{-2} d^{-1}, Thornton et al., 2016; 2020) as they have the greatest yet reported CH_4 concentrations in surface waters (e.g., 100 times above equilibrium, Shakhova et al., 2010). In the wintertime, there are also large gas bubbles trapped within the sea ice, and bubbles presumably consist of CH_4, but ice–air fluxes have not been measured. Several observations of under–ice CH_4 concentrations in different parts of the AO (Kvenvolden et al., 2003; Thornton et al., 2016) speculate that the CH_4 flux into the atmosphere is a seasonal feature occurring as a one–time event when the ice melts or breaks as in the case of smaller shallower northern lakes (e.g., Engram et al., 2020). However, this is obviously not the case for the dynamic and mobile pack ice. Flux from ice–covered but fractured AO areas in the Chukchi and East Siberian seas, the areas, which are close to geological CH_4 sources, has been reported to be relatively high in summer when ice concentrations decrease due to ice melt (2 mg CH_4 m^{-2} d^{-1}, Kort, et al., 2012), implying that sea ice dynamics and fracturing could play a significant role in the AO becoming a larger marine source of CH_4 into the atmosphere than previously estimated (e.g., Parmentier et al., 2015). Moreover, as shown in this study, newly formed sea ice in winter also emits CH_4 into the lower atmosphere. This puts emphasis on the importance of studies of CH_4 dynamics in sea ice, also in winter when the ice concentration is high and fracturing of the ice pack and subsequent new ice formation can result in increased potential for CH_4 evasion to the atmosphere.

4. Conclusions

We observed methane (CH_4) dynamics in under–ice water and new thin sea ice in the Nansen Basin of the Arctic Ocean (AO) following a winter storm. The many new fractures in the ice pack, initially areas of open water leads became consequently large areas of new thin and permeable sea ice, formed as a result of this storm (similar to that observed for a later storm the same winter (Itkin et al., 2018). During storm–induced ice break up, CH_4 vented into the air from supersaturated under–ice water (8–12 nmol L^{-1}) in open water leads (up to 30% of overall surface area) with a maximum flux of 1.04–2.13 mg CH_4 m^{-2} d^{-1}. Initially, newly formed sea ice in the leads was CH_4 supersaturated with respect to the atmosphere (5–7 nmol L^{-1}). During five days of observations, 2–3 nmol L^{-1} of this CH_4 escaped into the atmosphere until concentrations equilibrated with the atmosphere, and the ice became less permeable. This implies that the winter ice pack is not an impermeable barrier for CH_4 loss to the atmosphere, and not only the open water leads but also the sea ice itself plays an active role in this wintertime flux.

Understanding of CH_4 dynamics and associated processes in different sea ice conditions as well as under various meteorological events becomes an essential link for better estimates of CH_4 emissions from the CH_4 supersaturated AO surface waters and sea ice into the atmosphere. Sea ice is entering a new state from being largely thicker multi–year sea ice to predominantly thinner first–year thinner sea ice (Maslanik et al., 2011; Stroeve et al., 2012; Meier et al., 2014). Moreover, increasing the mean speed and deformation rate of the Arctic sea ice (Spreen et al., 2011), and rising frequency of winter storms and warming events in the Arctic (Graham et al., 2017; 2019) lead to an increasing amount of occurring fractures and open water leads. All these factors in addition to decreasing sea ice concentration in the AO, may enhance gas transfer intensity similar to what has been shown for CO_2 (Prytherch et al., 2017). The release of CH_4 into the atmosphere could be substantial in the future AO and is opposed to the scenario when CH_4 is majorly consumed by microbes while residing beneath sea ice cover (Kitidis et al., 2010). It is said that the CH_4 release rate from the East Siberian Sea estimated from atmospheric observations indicates that the bottom-up estimates could be overestimated (Tohjima et al., 2020). In–depth

multidisciplinary studies of changes in the coupled ocean–ice–atmosphere system with a focus on CH_4 dynamics and exchange will shed light on whether the AO itself is a more significant source of atmospheric CH_4 than previously thought (Myhre et al., 2016).

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Figure captions

Figure 1. (A) Drift track (red line) of R/V Lance during the N–ICE2015 campaign in January and February 2015, when it was anchored to Floe 1. White circles are positioned where ice coring in the lead took place. The background is from Copernicus Sentinel–1 satellite imagery (May 25, 2015; courtesy of European Space Agency) to indicate typical sea ice conditions in the study area.

Figure 2. Meteorological conditions (air temperature, air pressure, and wind speed) during the storm event and ice coring for this study. Periods of storm event and ice coring were also indicated.

Figure 3. Radar images of the sea ice surrounding R/V Lance, leads are areas with smooth black color while older sea ice with a rougher surface is shown by brighter shades taken from the ship–

based radar (Haapala et al., 2017), on (A) February 7 and (B) February 9. The radar images are about 7 km across. (C) Photo of the frost flowers covered sea ice in the lead taken on February 9. A meter–stick in the photo is for scale.

Figure 4. Relationship of k660 to u10 based on parametrizations from Wanninkhof (2014) (W14), Butterworth and Miller (2016) (B&M 16), and Prytherch (2020) (Pr20). Owfr5 and Owfr30 are for open water fractions of 5 and 30% respectively, representing minimum and maximum open water fraction during 12 days of this study. u is the measured wind speed at the meteorological mast at 10 m above the sea ice surface, which ranged between 0 and 25 m s^{-1}.

Figure 5. Methane concentrations in under–ice water sampled on (A) February 2, (B) February 3, (C) February 9, and (D) February 10. Water on all these days was sampled from under sea ice and not from the ice edge.

Figure 6. (A) Sea ice salinity, (B) ice temperature, and (C) and brine volume fraction for ice core of C1–C21 (Table 1 for reference). The light blue background shows the sea ice thickness. Grey in (C) indicates values equal or below 5%, which is a threshold for sea ice permeability.

Figure 7. (A) Sea ice CH_4 concentration and (B) CH_4 concentration to salinity ratio for ice core of C1–C21 (Table 1 for reference). The light blue background shows the sea ice thickness.

Figure 8. Calculated sea–to–air fluxes of CH_4 (top panel) using a surface CH_4 concentration of 10 nmol L^{-1}, with three different k660 parametrizations as in Table 2, bold font. Open water fractions in percent (bottom panel).

Table 1. The list of sea ice cores collected in the leads, with dates, exact coordinates for each date, ice thickness (cm), average ice salinity, average ice temperature (°C), average CH_4 concentrations (nmol L^{-1}), air temperature (T, °C), wind speed (m s^{-1}), sea level pressure (hPa), number of section.

Date	Core ID	Time (UTC)	Ice thickness (cm)	Average salinity	Average ice T (°C)	Average CH_4 (nmol L^{-1})	Air T (°C)	Wind speed (m s^{-1})	Sea Level Pressure (hPa)	Number of sections
7 Feb. 2015	C1	14:04	8	16.841	–4.6	6.2	–28.1	10.9	988	1
82.50 N	C2	14:14	8	15.347	–4.6	6.1	–28.2	10.7	988	1
17.81 E	C3	14:28	8	17.864	–4.6	6.7	–28.2	11.1	988	1
	C4	14:38	8	18.028	–4.6	7.1	–28.2	12.2	988	1
	C5	14:48	8	14.663	–4.6	7.2	–28.2	11.3	988	1
	C6	14:56	8	15.654	–4.6	5.4	–28.1	12	988	1
9 Feb. 2015	C7	19:48	18	14.649	–14.1	7.6	–35.6	4	982	1
82.34 N	C8	19:56	17.5	15.385	–14.1	5.6	–35.4	5.3	982	1
18.39 E	C9	19:57	18	15.625	–14.1	4	–35.4	5.1	982	1
	C10	20:10	17.5	16	–14.1	4.4	–35.5	5.2	983	1
	C11	20:19	16.5	14.873	–14.1	4.8	–35.6	4.7	982	1
	C12	20:34	17.5	15.407	–14.1	5.6	–35.6	5.1	982	1
10 Feb. 2015	C13	14:24	20.8	15.837	–11.8	4.5	–35.8	3.7	985	2
82.26 N	C14	14:31	21.4	16.176	–11.8	5.3	–35.7	5.2	985	2
18.79 E	C15	14:32	20	14.04	–14.15	5.4	–35.7	4.7	985	2
	C16	14:39	21.2	15.367	–11.8	3.8	–35.7	4.1	984	2
	C17	14:49	21	15.367	–11.8	3.8	–35.7	5.2	984	2
	C18	15:09	21.3	15.549	–11.8	7.5	–35.7	4.2	984	2
12 Feb. 2015	C19	14:46	28	13.144	–11.1	5.2	–35.7	3.2	985	3
82.09 N	C20	15:20	27	11.803	–11.1	4.9	–36.0	4.5	984	3
19.25 E	C21	15:35	27	12.162	–11.1	4.1	–35.5	5.2	984	3

Table 2. Different k660 parameterizations.

Parameterization (k_{660} (cm hr^{-1})); u (m s^{-1})	Source
$0.251 \cdot u^2$	Wanninkhof (2014)
$0.245 \cdot u^2 + 1.3$	Butterworth and Miller (2016)
$0.189 \cdot u^2$	Prytherch and Yelland (2021)
$0.251 \cdot u^2 \cdot f$	Wanninkhof (2014) scaled to f – open water fraction

Table 4. Sea–to–air fluxes (mg CH_4 m^{-2} d^{-1}) in different areas of the AO. All values are positive (flux from the ocean to atmosphere).

Source	Flux	Latitude	Longitude	Season or month	Sea ice conditions	Location/region	Method used
This study	0.23	82.08	19.25	winter (Feb)	ice covered with leads open	Nansen basin	Calculated based on Wanninkhof 2014
Shakhova et al. (2010)	3.67	75.04	128.74	summer	ice–free	Laptev Sea (mean)	Calculated based on Wanninkhof 1992
Thornton et al. (2016)	3.8	73.16	166.1	summer	ice–free	Western East Siberian Sea	Calculated based on Wanninkhov 2014
Thornton et al. (2020)	1.5			summer	Mixed ice covered/ice free	Laptev+East Siberian+Chukchi seas	Eddy covariance measurements
Kort et al. (2012)	2	82.53	145	Nov, Apr	ice covered with leads open	North of Alaska	Estimations based on air mole fraction ppm and eddy diffusivity 0.3 m^{-2} s^{-1}
He et al. (2013)	0.56	86.81	173.24	summer	ice–covered	Central Arctic Ocean	Chamber technique measurements
Damm et al. (2007)	1.05	77.31	19.35	Mar	polynya	Storfjorden Polynya	Calculated based on Wanninkhof 1992
Silyakova et al. (2020)	0.24	78.38	10.48	summer	ice–free	West Spitsbergen	Calculated based on Wanninkhof 2009
Graves et al. (2015)	0.32	78.55	9.42	summer	ice–free	West Spitsbergen	Calculated based on Wanninkhof 2009
Lammers et al. (1995)	0.08	74.9	27.56	Aug	ice–free	Barents Sea	Calculated based on Wanninkhof 1992

Table 3. CH_4 fluxes calculated with different k660 parametrizations. Fluxes are calculated as maximum, minimum, and mean values for the storm event (3–8 February) and post–storm low winds.

		Storm event (winds >7 m s^{-1})			Still, no storm (winds <7 m s^{-1})		
		8 nmol L^{-1}	10 nmol L^{-1}	12 nmol L^{-1}	8 nmol L^{-1}	10 nmol L^{-1}	12 nmol L^{-1}
k_{660} W14	Max	1.04	1.59	2.13	0.09	0.13	0.18
	Min	0.01	0.02	0.02	0.02	0.03	0.03
	Mean	0.2	0.31	0.41	0.05	0.08	0.1
k_{660} Pr20	Max	0.79	1.2	1.6	0.07	0.1	0.14
	Min	0.01	0.01	0.02	0.01	0.02	0.03
	Mean	0.15	0.23	0.31	0.04	0.06	0.08
k_{660} W14OW fr	Max	0.1	0.15	0.2	0.02	0.03	0.05
	Min	0	0	0.01	0	0	0
	Mean	0.03	0.04	0.06	0	0.01	0.02