

Air-lake exchange of methane during the open water season in Syowa Oasis, East Antarctica

SASAKI, Masafumi¹, Noboru ENDOH¹, Satoshi IMURA², Sakae KUDOH², Takashi YAMANOUCHI³, Shinji MORIMOTO³ and Gen HASHIDA³

1. Department of Mechanical Engineering, Kitami Institute of Technology, Japan

2. Bioscience Group, National Institute of Polar Research (NIPR), Japan

3. Meteorology and Glaciology Group, National Institute of Polar Research (NIPR), Japan

Abstract

Dissolved methane concentrations (DM) in 17 lakes were measured as a part of the operations of the 45th Japanese Antarctic Research Expedition in ice-free rocky areas along the eastern coast of Lützow-Holm Bay (Syowa Oasis) in East Antarctica in the summer of 2003~2004. DM at the surfaces of 14 lakes ranged from the atmospheric equilibrium concentration (about 4 nmol L⁻¹ for fresh water) to 385 nmol L⁻¹. Relatively low DM of less than 50 nmol L⁻¹ were observed at about 60% of the lakes. Many of the lakes (area fraction of 85%) were supersaturated and are thus sources of methane to the atmosphere. The exchange coefficient was calculated using wind speed data at Syowa Station. Area fraction frequency distributions with 4 surface DM ranges were applied to all lakes at Syowa Oasis (110 lakes: total lake area of 9 km²). Extrapolation to the whole Syowa Oasis gives an estimate of total emission of about 1 t-CH₄ yr⁻¹. This is the first estimation of methane flux from the surfaces of thawed lakes to the atmosphere in Antarctica. Since methane efflux of about 2 t-CH₄ yr⁻¹ from frozen bubbles in lake ice was estimated in our previous study, a total amount of 3 t-CH₄ yr⁻¹ would be released to the atmosphere from the lakes during the ice melting season (December ~ January) at Syowa Oasis.

1. Introduction

It is well known that there are many lakes that maintain liquid water even in winter in Antarctica, and it is also known that there are microbial activities even in perennially ice-covered lakes. Dissolved methane concentrations (DM) in a lake's water column reflect microbial activities. *Wand et al.* [2006] reported an extremely high DM of 21.8 mmol L⁻¹ at Lake Untersee, an oligotrophic, perennially ice-covered freshwater lake in central Dronning Maud Land. It is thought that such high accumulation of DM is caused by the perennial ice cover that prevents convection and gas exchange between water and the atmosphere. *Andersen et al.* [1998] found a relatively low DM level of 63-156 nmol L⁻¹ near the bottom (anaerobic zone) of a perennially ice-covered amictic lake, Lake Hoare in McMurdo Dry Valleys. Lake Hoare receives water and sediment from glacial melt. *Smith et al.* [1993] showed that methane concentrations were < 1 μmol L⁻¹ in the upper oxic waters but increased below the oxycline to 1 mmol L⁻¹ in an amictic, permanently ice-covered lake, Lake Fryxell, separated by Canada Glacier only 3 km from Lake Hoare.

On the other hand, seasonally ice-covered amictic lakes are frequently found in coastal areas and islands, such as Livingston Island in South Shetland Islands [*Toro et al.*, 2007], King George Island and James Ross Island [*Hansson et al.*, 1992] near the Antarctic Peninsula and lakes with no outflow in Schirmacher Oasis [*Ramaiah*, 1995]. Microbial activities and methane have generally been found, regardless of trophic status, in freshwater lakes on Signy Island, South Orkney Islands [*Ellis-Evans*, 1982 and 1984]. *Ellis-Evans* [1984] reported a high DM of 5.7 μmol L⁻¹ at 0.1 m

above the sediment surface in the winter anoxic zone. With the disappearance of ice cover, DM in sediment (at 1 cm beneath the sediment surface) decreased steadily. They are similar to the lakes in Syowa Oasis as reported in the present study. DM data at the surface of seasonally ice covered lakes in Antarctica, however, could not be found in the literatures.

There are several ice-free rocky areas, the so-called Syowa Oasis, along the eastern coast of Lützow-Holm Bay (Soya Coast) in East Antarctica. More than one hundred lakes are located in these areas and the lakes have no ice cover during summer (December and January) [Yoshida *et al.*, 1975]. Only vegetation of mosses and algae is usually found on the bottom of these freshwater lakes [Imura *et al.*, 1999], though benthic copepods were found and they were first sampled at one of the melomictic lakes (Lake Nurume Ike) in the Langhovde area in 2007 [Kudoh *et al.*, 2008]. The staff of the National Institute of Polar Research (NIPR) has been carrying out the REGAL (Research on Ecology and Geohistory of Antarctic Lakes) Project to study biological activities in polar lakes since the 36th Japanese Antarctic Research Expedition (JARE36; 1994-1996). Since there is no inflow or outflow of organic materials in Antarctic lakes, the carbon cycle in freshwater lakes is very simple. Air-lake exchange is an important process for carbon balance in such a closed system.

The staff of NIPR also has been conducting the Atmospheric Circulation and Material Cycle in the Antarctic Program [Yamanouchi *et al.*, 1999], and in situ observations of atmospheric trace elements such as CO₂, CH₄, CO and O₃ and air samplings have been carried out at Syowa Station since 1984. From the viewpoint of atmospheric science, it is necessary to quantitatively evaluate the impact of methane efflux from lakes on atmospheric concentrations at the monitoring station in Syowa Station and also on global methane budget. Two different processes of methane efflux (or exchange) have been observed in seasonal ice-covered lakes in Syowa Oasis. One process is exchange between air and the lake water surface caused by chemical potential difference, and the other is direct efflux from bubbles suspended in lake ice early in the melting season. Sasaki *et al.* [2009] estimated the flux of methane from ice bubbles in Syowa Oasis. Though another possible process was ebullition from unfrozen lakes, no bubbling was found perhaps because of insufficient observations with a bubble trap.

The present study was undertaken to estimate the flux of methane from open water in seasonally ice-covered lakes. Observations of DM in lake water were carried out by JARE45 in summer (December 2003~January 2004) in the Syowa Oasis, East Antarctica.

2. Site description

The lakes studied are located in Syowa Oasis, along the eastern coast of Lützow-Holm Bay at 39°15'~ 45' E and 69°00'~ 45' S as shown in Fig. 1. Syowa Station was established by Japan in 1957 on East Ongul Island in the northernmost part of this area. Syowa Oasis consists mainly of the Ongul Islands [Ishikawa *et al.*, 1994], Langhovde [Ishikawa *et al.*, 1976], Skarvsnes [Ishikawa *et al.*, 1977] and Skallen [Osanai *et al.*, 2004]. The Lützow-Holm bay region is exposed by various kinds of metamorphic rocks, including pyroxene gneiss and garnet gneiss, without organic sedimentary rocks. Raised beach deposits, which can be found up to 20 m above sea level along the coastal region, result from isostatic uplift following the Last Glacial Maximum [Miura *et al.*, 1998, 2002]. Locations of the lakes studied in the Langhovde area and the Skarvsnes area are shown in Fig. 2 and Fig. 3, respectively. Almost all lakes (more than one hundred) have no ice cover for 1.5~2.5 months (no sunset season for 1.5 months from December to January) but are completely covered with ice for the ten months. Since the depths of most of the lakes (2~32 m) are greater than the maximum ice thickness (1.7 m), the lakes have liquid water under the ice cover even in the coldest season. On the bottoms of some lakes, therefore, vegetations of mosses and algae are still active even during winter. The presence of 'moss pillars' in Lake Hotoke Ike and 'pinnacles' in Lake Namazu Ike are impressive evidence of abundant vegetation in spite of highly oligotrophic states [Kudoh *et al.*, 2009].

On the other hand, there are smaller colonies of mosses and algae (no plants, no animals) on land sporadically. Since there are generally no inflows except melted water of snow or glaciers, no exchange of organic materials with

the outer field occurs in these lakes. The existence of geologic emission of methane from deep gas seepage was thought to be unlikely because DM from seawater was never found around the coasts in Syowa Oasis [Sasaki *et al.*, 2005]. Therefore, these lakes provide unique, simple ecosystem.

The lakes in Syowa Oasis are divided into four types [Seto *et al.*, 2002]. Type 1 lakes receive water and sediments from glacial melt. Salinities are generally less than 0.02 psu. Type 2 lakes (0.02~0.1 psu) receive water from glacial melt indirectly. Only water from snow melt comes into Type 3 lakes (0.1~2 psu). Algal debris can be seen in both Type 2 and Type 3 lakes. Type 4 lakes have been isolated from the ocean by uplift due to deglaciation in the Holocene and they have seawater-origin water. The maximum elevation of Type 4 lakes is Lake Kobachi Ike, the water surface level of which is 25 m in altitude. Carbon-14 AMS (Accelerator Mass Spectrometry) dating (AMS^{14}C) of sediment cores showed that lakes at an altitude of less than 8 m were isolated from the ocean later than 3500 years before present (yBP) (corrected by only $\delta^{13}\text{C}$) [Seto *et al.*, 2002].

The thicknesses of sediments at the lake bottoms ranged from 55 cm to 2 m. In some lakes, most of the sediment consisted of thick algal debris (sometimes more than 1 m in thickness, such as in Lake Skallen O-Ike). In other lakes, for example, Lake Maruwan O-Ike, half of the thickness of sediment was old oceanic sediment that contained oceanic Foraminifera. Sedimentation rates were calculated to be 0.17~0.67 mm yr⁻¹ for several lake cores [Matsumoto *et al.*, 2006]. High total organic carbon contents (TOC) were found in recent sediments that had formed after isolation from the ocean. The core top age was 300~400 yBP.

These site characteristics mentioned above suggest that the methane origin is limited as follows.

- 1) There is not neither biogenic nor thermogenic methane production in the rocks, because Syowa Oasis is exposed by only metamorphic rocks without organic sedimentary rocks.
- 2) The existence of geologic emission of methane from deep gas seepage was thought to be unlikely because DM from seawater was not found around the coasts in Syowa Oasis [Sasaki, 2005].
- 3) There is no (or very small) inflow of organic matter from outer field.

It can therefore be assumed that methane is produced exclusively by microbial activity within recent lake sediments. Complete compositional analyses of dissolved gases and isotopic analysis of CH₄ are however necessary to completely understand the methane formation process and origin.

3. Methods

3.1 Water sampling and DM analysis

Each location of water sampling in each lake was chosen at the point of maximum depth of the lake. Surface water temperature (0~5 cm deep) and sample water temperature were measured by a K type thermocouple. Water samples were obtained by using a Kitahara water sampler (1 L) ([http://at2.tactnet.co.jp/rigo/dldata/1WaterBottle.pdf#search=Kitahara water sampler](http://at2.tactnet.co.jp/rigo/dldata/1WaterBottle.pdf#search=Kitahara%20water%20sampler)). The water samples were poured calmly with a rubber tube in 500 mL glass vials, which was washed by strong shake with the sample water of 100~200 mL at first. After 200~300 mL of the sample was spilled (overflow procedure), the vial was sealed. The samples were preserved in a refrigerator (around 5 °C) in the icebreaker 'Shirase' on the return cruise to Japan and they were analyzed in Kitami Institute of Technology. Since the maximum preservation period was five months, HgCl₂ was added as a preservative to the water samples. HgCl₂ concentration after poisoning was 0.4 μmol L⁻¹. Even with the addition of a relatively high concentration of HgCl₂, DM may decrease during transportation to Japan. Seawater samples (as a typical oligotrophic water) which contained the preservative with the same concentration and DM of 1 μmol L⁻¹ adjusted by bubbling of pure methane gas were used for DM reduction calibration. DM decreased 15% after one month and 18% after three months. There was no correction of the decomposition (or pass through a vial cap) of DM in this study.

A headspace technique was used to measure DM in water. A part of the water sample in each of three glass vials (27 mL each) was replaced by pure nitrogen, leaving 10 mL of water sample in each vial. After each vial sample had

been heated and kept at 60 °C for more than 20 minutes, 1 mL of headspace gas was injected into a gas chromatograph equipped with a flame ionization detector (GC-FID) which had a 3-meter-long stainless column filled with Porapak-Q (http://www.daichem.co.jp/eng/products/capillary_c/gs_q_en.html). Nitrogen was used as the carrier gas. Standard gas (CH₄ of 2.026 ppmv balanced with N₂) injected into a vial of the same size with 10 mL of deionized water with boiling procedure was used as a span gas. The minimum detectable DM by this technique was about 15 nmol L⁻¹. The analytical errors were within ±5%.

3.2 Estimation of methane flux from all lakes in Syowa Oasis

Notation

Variable

A	total (110) lake area in Syowa Oasis (= $8.92 \times 10^6 \text{ m}^2$)
C	gas concentration (mol m ⁻³)
D	diffusion coefficient (m ² s ⁻¹)
DM	dissolved methane concentration (mol m ⁻³)
DM ₀	atmospheric equilibrium concentration of dissolved methane (mol m ⁻³)
f_m	mass flux density (g-CH ₄ m ⁻² hr ⁻¹)
f_{MOL}	mol flux density (mol-CH ₄ m ⁻² hr ⁻¹)
H	Henry's coefficient
k_{600}	transfer coefficient of CO ₂ in freshwater in Sc of 600 at 20 °C (m s ⁻¹)
k	transfer coefficient (m s ⁻¹)
m	total mass flux (g-CH ₄ yr ⁻¹)
Sc	Schmidt Number
Δt_A	period of open water season = two months = 1440 hr
t_s	water temperature at the lake surface (°C)
M	molecular weight (g mol ⁻¹)
U	wind speed (m s ⁻¹)
U_{10}	equivalent wind speed at a height of 10 m (m s ⁻¹)
z	area fraction (m ² m ⁻²)
ν	kinematic viscosity (m ² s ⁻¹)

Suffix

a	bulk air
CH ₄	methane
i	DM classification (i=1~4)
s	surface
b	bottom
w	bulk water

The concept of gas exchange between water surface and the atmosphere was shown as the two-film model [Liss and Slater, 1974]. Exchange flux is proportional to gas concentration difference between water film and air film at the boundary as follows.

$$\text{flux} = k (C_w - C_a/H) \quad (1)$$

Where k is transfer coefficient (= transfer velocity), C_w is the concentration in the bulk water, C_a is the gas

concentration in the bulk air and H is Henry's coefficient. For low solubility gases such as CO_2 and CH_4 , in practice, the liquid phase coefficient can be regarded as k , because the rate limiting step is transfer through the water side film as molecular diffusion through water is considerably slower than in air. In Eq. (1), positive flux means release of gas from water to the atmosphere. Since k behaves strong and nonlinear dependence on wind speed, enormous theoretical and experimental studies have been made to determine k . Results of studies on gas exchange between oceans and the atmosphere have been summarized well by *Nightingale and Liss* [2003]. Recent important topics on gas transfer at water surface were introduced by *Donelan et al.* [2002]. Many researchers have focused on the exchange of CO_2 , which is regarded as one of the most important greenhouse gases. Although there have not been many studies on air-lake exchange of methane, information on air-sea exchange of CO_2 is available for estimation of methane flux. Several empirical formulations express a relationship between exchange coefficient k and wind speed U . The formula proposed by *Sebacher et al.* [1983] was obtained by a direct measurement of CH_4 flux in a wind chamber with floats on lake surface in relatively low wind speed range, as follows.

$$k_{\text{CH}_4} = 4.72 \times 10^{-6} \quad (\text{m s}^{-1}) \quad (U = 0) \quad (2)$$

$$k_{\text{CH}_4} = 3.06 \times 10^{-6} + 3.33 \times 10^{-6} U^{1.96} \quad (\text{m s}^{-1}) \quad (U = 1.4 \sim 3.5 \text{ m s}^{-1}) \quad (3)$$

It was difficult to apply this formula to actual field, because the U was measured at 2 cm above the water surface. However, the fact that k_{CH_4} was not zero at $U=0$ must be retained.

Inert volatile tracers, particularly SF_6 , have been deliberately added to water bodies in order to determine k in recent empirical field studies. Dual tracer method usually using SF_6 and ^3He is required in order to correct for their dilution due to horizontal and vertical mixing. *Wanninkhof* [1992] proposed a simple parabolic relationship, Eq. (4), between k and U , from the dual tracer observation. *Solomon et al.* [2009] applied the equation to air-sea exchange of methane near methane bubble plumes in the Gulf of Mexico.

$$k_{600} = 0.862 \times 10^{-6} U^2 \quad (\text{m s}^{-1}) \quad (4)$$

k_{600} is the transfer coefficient of CO_2 in freshwater in the Schmidt number Sc of 600 at 20 °C. For methane exchange the Schmidt number for methane, Sc_{CH_4} , has to be used to obtain the correct gas exchange coefficient k_{CH_4} ,

$$k_{\text{CH}_4} = k_{600} (Sc_{\text{CH}_4} / 600)^{-n} . \quad (5)$$

The exponent $n = 0.5$ was observed in many field experiments under a higher wind speed condition ($U_{10n} \geq 3.6 \text{ m s}^{-1}$). The value was also supported by theoretical considerations for surface renewal model (*Ledwell*, 1984) and eddy models (*Fortescue and Pearson*, 1967, *Lamont and Scott*, 1970).

Cole and Caraco [1998] proposed a formulation for air-lake exchange of CO_2 under lower wind condition from measurements using SF_6 addition.

$$k_{600} = 5.75 \times 10^{-6} + 0.597 \times 10^{-6} U_{10}^{1.7} \quad (\text{m s}^{-1}) \quad (U_{10} = 0.5 \sim 9.5 \text{ m s}^{-1}) \quad (6)$$

Eq. (6) also show that k_{600} is not zero at $U_{10}=0$. The equation was compared with the following formula obtained by *MacIntyre et al.* [1995].

$$k_{600} = 1.25 \times 10^{-6} U_{10}^{1.64} \quad (\text{m s}^{-1}) \quad (7)$$

Cole and Caraco [1998] concluded the equation fitted the observations well, including the low-wind end.

Nightingale et al. [2000] proposed an approximation, Eq. (8) as the best fit for three dual tracer datasets for the ocean obtained by *Wanninkhof* [1992], *Smethie et al.* [1985] and *Liss and Merlivat* [1986] as

$$k_{600} = 0.64 \times 10^{-6} U_{10}^2 + 0.28 \times 10^{-6} U_{10} \quad (\text{m s}^{-1}) \quad (8)$$

Nightingale et al. [2000] also proposed another approximation in the same paper as follows,

$$k_{600} = 0.617 \times 10^{-6} U_{10}^2 + 0.925 \times 10^{-6} U_{10}. \quad (\text{m s}^{-1}) \quad (9)$$

Eq. (9) gives a best fit to 9 dual tracer datasets (including 3 datasets above and additional 4 datasets in the North Sea of *Nightingale et al.*, 2000, Georges Bank data of *Asher and Wanninkhof*, 1998 and Florida Shelf data of *Wanninkhof et al.*, 1997). Both Eq. (8) and Eq. (9) are valid for wind speed conditions of $\sim 15 \text{ m s}^{-1}$.

The transfer coefficient was computed using the empirical *Nightingale* relationship Eq. (9) [*Nightingale et al.*, 2000] in this study at first. The result was compared with results obtained by using other relationships described above in the Discussion section.

Hourly mean wind speed observed at the meteorological observatory in Syowa Station was directly applied as U_{10} in Eq. (9) during the period from December 2003 to January 2004, because the sensor height was 10.1 m. Although Langhovde, Skarvsnes and Skallen areas are located about 30 km, 60 km and 90 km, respectively, away from Syowa Station (Ongul Islands), they are within the same meteorological range. The wind speed data at Syowa Station were therefore applied to the whole Syowa Oasis.

In the present study, since the period in which hourly mean wind speed exceeded 15 m s^{-1} was only 3% (maximum wind speed: 33.1 m s^{-1}) of the total open water season (about 2 months), stronger U dependence ($\sim U^3$) [*Wanninkhof et al.*, 1999] on k because of white caps (wave breaking) and bubble exchange was not considered.

When k_{CH_4} is determined, methane flux can be calculated using difference of DM at the water surface and corresponding atmospheric equilibrium concentration (DM_0) as follows.

$$f_{\text{MOL}} = k_{\text{CH}_4} (\text{DM} - \text{DM}_0). \quad (10)$$

The lakes were divided into 4 classes by their surface DM levels. Area fraction z (=lake area / total area of 14 lakes) for each class was obtained. Assuming that the area fraction frequency distributions to DM ranges for 14 lakes surveyed can be applied to the whole Syowa Oasis, including 110 lakes, total mass flux of methane m in Syowa Oasis would be given by the following equations.

$$m = \sum_{i=1}^4 (f_{m,i} z_i A \Delta t_A) = A \Delta t_A \sum_{i=1}^4 (f_{m,i} z_i). \quad (11)$$

$$f_m = M_{\text{CH}_4}^{-1} f_{\text{MOL}}. \quad (12)$$

4. Results

4.1 DMs in open lake water

The results of DM analysis in 17 lakes during the ice melting season from December 2003 to January 2004 are shown in Table 1. The abbreviation ND means not detectable level of DM (less than 15 nmol L^{-1}). The number in bracket after each name of lake in Table 1 shows lake type mentioned in Site description section.

As mentioned above (Site description section), it can be assumed that methane is produced exclusively by microbial activity within lake sediments. It therefore seems reasonable that DM (both at depths of 1 meter and 4 meters) in Lake Tennno-kama Ike, a Type 1 lake, are ND level because of low temperature due to contact with a

glacier front. Another ND at the surface was unexpectedly found in Lake Akebi Ike, a meromictic lake (Type 3 or Type 4), in which abundant vegetation was expected. The maximum surface DM was 385 nmol L^{-1} in Lake Oyako Ike (Type 3). Surface DM exceeds 100 nmol L^{-1} in only 30% of lakes. Water columns in freshwater lakes are well mixed during the open water season [Kudoh *et al.*, 2009]. Relatively large positive dependence of DM on depth, however, was observed at Lake Yukidori Ike (Type 2 or Type 3), a freshwater lake. The reverse (negative) dependence was also found at Lake Kamikama Ike (Type 2 or Type 3).

On the other hand, in the ice-cover season, high DM anomalies were frequently found near the bottoms of the lakes, e.g., DM of more than $1 \text{ } \mu\text{mol L}^{-1}$ was found in 60% of the lakes studied in the Skarvsnes area [Sasaki *et al.*, 2009]. Dissolved methane accumulates near the bottom of freshwater lakes due to anaerobic microbial activity in the sediment, and also due to no (or very weak) convection during the ice-cover season. At the beginning of ice melt, DM near the bottom of the lake is transported to the surface mainly by wind-induced convection. This suggests that surface DM will change with a time lag after ice melt. There is no correlation between DM near the bottom (DMb: 0.5 m above the lake bottom) during the ice-cover season (the data by Sasaki [2009]) and the surface DM (DMs) during open water season (hollow circles), while positive correlation (dotted line) between the DMb and methane concentrations in frozen bubbles in the ice cover can be recognized (solid circles) as shown in Fig. 4. One of the reasons of such uncorrelated behavior between DMb and DMs may be data of various time lag after ice melt for 2 months. Phelps *et al.* [1998] reported that the highest level of methane efflux from open water was observed during a brief period just after ice melt in high latitude lakes.

The maximum DM of $21 \text{ } \mu\text{mol L}^{-1}$ in the open water season was found at deeper (9 m deep) water in Lake Kobachi Ike, a saline lake (Type 4). Salinity was not so high (16.5 psu) and dissolved oxygen concentrations (DO) were almost at equilibrium concentration (11.9 mg L^{-1}) even at that depth. Franzmann *et al.* [1991] reported extremely high DM of 4.9 mmol L^{-1} near the bottom of Ace Lake in Vestfold Hills, Antarctica. Salinity in the saline layer (below 10 m) was similar to that of seawater and DM was undetectable (less than $0.9 \text{ } \mu\text{mol L}^{-1}$) in the shallower layer. In hypersaline lakes such as Lake Suribachi Ike and Lake Funazoko Ike which have higher salinities than 200 psu, extremely low DO and strong smell of H_2S at the deeper water column than the oxycline are always observed. Although anomalous DM were also found below the oxycline, surface DM was not so high in those hypersaline lakes, perhaps because of strong stratification. There are three layers with two oxyclines in Lake Suribachi Ike, and the highest DM was found in the middle layer (at 11 m) as shown in Table 1. Such hypersaline lakes have also been found in other areas in Antarctica (Vestfold Hills [van den Hoff and Franzmann, 1986] and McMurdo Oasis [Boswell *et al.*, 1967], etc).

Atmospheric equilibrium DM ($=\text{DM}_0$) depends on water temperature, salinity of water, atmospheric pressure and concentration of methane. DM_0 was calculated using the approximation by Wiesenburg *et al.* [1979]. DM_0 are 4.6 and 3.9 n mol L^{-1} at surface water temperatures of 0 and $5 \text{ }^\circ\text{C}$, respectively, under conditions of 1 atm, 1.8 ppmv- CH_4 and 0 psu. When the surface DM exceeds DM_0 , the lake acts as a methane source to the atmosphere.

Only 14 DM data records (underlined values in Table 1) can be used as the surface DM. They were classified into 4 classes by the surface DM range ($i=1$: ND, $i=2$: equilibrium~50, $i=3$: 50~100, $i=4$: greater than 100 nmol L^{-1}). ND was regarded as the equilibrium DM (no exchange). Areal weighted mean DM in each class is shown in Fig. 5 (circles). Lake area fraction in each class z_i is also shown in Fig. 5 (bars). The number in each bar indicates the number of lakes surveyed. The area fraction of $i=2$ was the largest, about 60%, and the average DM ($=\text{DM}_2$) was $24.2 \text{ n mol L}^{-1}$. The surface DM exceeded DM_0 in 85% of the lake area.

4.2 Exchange of methane between the atmosphere and lake surface

Equation (9) [Nightingale *et al.*, 2000] shows nonlinear dependence of exchange coefficient k_{600} on wind speed U_{10} . Averaging of wind speed must therefore be done carefully. k_{CH_4} computed by Eq. (9) using both hourly mean wind speed and daily mean wind speed are shown in Fig. 6. Estimation of total methane flux using hourly mean

wind speed was about 14% greater than that using daily mean wind speed. The results using hourly mean wind speed are shown below.

k_{CH_4} also depends strongly on surface water temperature (on Sc_{CH_4}). Diffusion coefficient of DM (in water) D increases and kinematic viscosity ν decreases with temperature rise. As the result, $Sc (= \nu/D)$ decreases sharply with temperature rise. When the surface water temperature rises from 0 to 5 °C, k_{CH_4} increases by about 9%. Water temperatures at lake surfaces (t_s) in Table 1 showed a range of 0.9~9.0 °C. Higher temperatures were frequently found in saline lakes (Type 4) due to the solar pond effect. Areal weighted mean temperature at the surface in 14 lakes was 2.7 °C. Total mass flux of methane released from open water lakes to the atmosphere in Syowa Oasis was estimated for surface water temperature of 2.7 °C ($Sc_{\text{CH}_4} = 924$, $DM_0 = 4.2 \text{ nmol L}^{-1}$) as follows:

$$0.417 \text{ (December 2003)} + 0.852 \text{ (January 2004)} = 1.269 \text{ (t-CH}_4 \text{ yr}^{-1}\text{)}.$$

The reason for the mass flux in January being about two-times that of December is the higher wind speed in January.

5. Discussion

Summer (especially December) is the calmest season in Syowa Oasis. The accumulated period in which hourly mean wind speed was less than 3.6 m s^{-1} was 49%. When alternative exponent of $n=2/3$ proposed by *Deacon* [1997] (boundary layer model) was applied for a calm water surface condition ($U_{10m} < 3.6 \text{ m s}^{-1}$) to Eq. (9), the total methane efflux was smaller by only 0.7% (December; -1.0%, January; -0.7%) than that at constant $n=0.5$.

Eq. (8) proposed by *Nightingale* gave 8% lower total methane release than that given by Eq. (9) (= the reference equation). Eq. (4) proposed by *Wanninkhof* [1992] gave 19% higher total methane than the reference. On the other hand, the total methane flux calculated from Eq. (7) proposed by *MacIntyre* [1995] was 33.7% lower than the reference. The approximation for air-lake exchange Eq. (6) by *Cole and Caraco* [1998] gave 3.6% higher flux than the reference, when the equation was applied under lower wind condition ($U_{10} < 3.6 \text{ m s}^{-1}$). Table 2 summarizes deviations of total mass flux of methane by various formulas from that by *Nightingale's* equation Eq. (9) as the reference. While the deviation is -33.7~+19%, each formula also has large scattering in itself.

There are several uncertainties elsewhere in this study as follows.

- 1) Samples were analyzed after long-term preservation (5 months max.) from sampling. This would bring uncertainty of about -20% in DM measurement. DM analysis at least within one week after sampling (at Syowa Station) is desired in the future studies.
- 2) The hourly mean wind speed was used for exchange coefficient estimation. This gave 14% larger estimation of k_{600} than that using daily mean wind speed. The higher time resolution of wind, the better, generally, because k nonlinearly depends on wind speed. However, when a proposed formula is applied to k estimation, the time resolution must be agreed with the time averaging wind speed which was used in the formulation of the equation. The total uncertainty summarized above including DM analysis and different k formulations (Eq. 4 ~ Eq. 9) was the total methane flux of -50% ~ +42%. The following uncertainties could not be estimated in this study.
- 3) The formulas from observations in the oceans except Eq. (6) were applied to the estimations of flux at lakes. Trees and other structures on land decelerate wind speed generally. Though there is no trees, bushes, grasses and any synthetic structures around lakes in Antarctica, some topographical obstructs must not be always negligible, e. g. a deep conical concavity at Lake Kamikama-ike. Shallower depth and smaller surface area in lakes than those in the ocean will make difference in wave (turbulence) formations. In spite of these difference between lakes and the ocean, the estimated flux by Eq. (6) (for lakes: *Cole and Caraco*, 1998) lied mid-value between the deviation of various formulas. However, *Eugster et al.* [2003] investigated air-lake exchange of CO_2 employing the eddy covariance method and concluded that significant differences between the governing physical exchange processed over lakes and the ocean may exist. *MacIntyre et al.* [2002] calculated gas transfer coefficient using

surface renewal model and concluded that the contribution of buoyancy fluxed to the gas transfer coefficient was very important under lower wind speed ($<5\text{-}6\text{ m s}^{-1}$) condition. The calculated coefficient was similar to estimation from *Cole and Caraco, 1998*.

- 4) A surface DM of a lake was represented by only one sample near center of the lake in this study. Actually, surface DM would distribute over water surface. Observations of DM distributions are required in order to make the estimations more accurate and also to understand topographical effects of lake bottom on solar radiation, on turbulence (depth effect) and so on.
- 5) Seasonal change in surface DM during open water season was not observed. Very high DM was expected in the early ice melting period [*Phelps et al., 1998*]. Time series observations during melting to freezing period is required in order to estimate the effect of accumulation of DM during ice covered season on methane flux.
- 6) Since observation of ebullition during summer was insufficient (only 5 days by a bubble trap), no ebullition was found during the open water season. *Walter et al. [2008]* often observed methane ebullition in all seasons of the year in Northern Siberia. They developed a smart procedure to observe ebullitions. When they found a bubble clusters in ice cover, they recorded the accurate location and set a bubble trap there during open water season. Ice cover is an ideal bubble trap during winter. *Sasaki et al., 2009* reported that the mean bubble volume was about 0.6% of ice volume, while the methane gas concentrations in bubbles varied of 5 orders of magnitude. About $2\text{ t-CH}_4\text{ yr}^{-1}$ (about two-times that of gas transfer by diffusion in this study) from frozen bubbles in lake ice was released early ice melt season in Syowa Oasis.

The uncertainties listed in 5) and 6) would cause conservative estimations of total methane release.

- 7) Wind speed observed by the observatory at Syowa Station was used for flux estimations in the whole Showa Oasis including an area 90 km away from Syowa Station. Continuous wind observations by at least one automatic weather station at each area of Langhovde, Skarvsnes and Skallen are desired.

In spite of these uncertainties, an estimate of total flux (from diffusion) of about $1\text{ t-CH}_4\text{ yr}^{-1}$ would be a good approximation in Syowa Oasis. Since methane efflux of about $2\text{ t-CH}_4\text{ yr}^{-1}$ from frozen bubbles in lake ice was reported by *Sasaki et al. [2009]*, a total amount of $3\text{ t-CH}_4\text{ yr}^{-1}$ would be released to the atmosphere from the lakes during ice melting season (December ~ January) in Syowa Oasis. This corresponds to an annual average flux of $0.34\text{ g-CH}_4\text{ m}^{-2}\text{ yr}^{-1}$. *Juutinen et al., 2009* estimated (using Eq. 7) an average methane flux from 177 boreal lakes in Finland of $0.78\text{ g-CH}_4\text{ m}^{-2}\text{ yr}^{-1}$ (about two-times that in Syowa Oasis). *Etioppe et al. [2004, 2008]* estimated that global geologic methane emission from deep seepage ranges from 43 to 64 Tg yr^{-1} (possibly approaching 80 Tg yr^{-1}). Emissions from individual seepage areas are in the order of $10^2\text{-}10^3\text{ g-CH}_4\text{ m}^{-2}\text{ yr}^{-1}$ for mud volcanoes and may reach the order of $10^5\text{ g-CH}_4\text{ m}^{-2}\text{ yr}^{-1}$ for large gas seeps, such as those producing "eternal flames" [*Etioppe, 2009*]. Microseepage, which is a diffuse exhalation of methane from soil, may range from a few units to thousands of $\text{mg m}^{-2}\text{ day}^{-1}$ [*Etioppe et al., 2008*]. Regarding biogenic methane emissions from thaw lakes, *Zimov et al. [1997]* and *Walter et al. [2006, 2007]* reported a total output of $1.5\text{-}3.8\text{ Tg-CH}_4\text{ yr}^{-1}$ for Northern Siberia, with an average mass flux of about $25\text{ g-CH}_4\text{ m}^{-2}\text{ yr}^{-1}$. The estimated Syowa Oasis areal emission of $3\text{ t-CH}_4\text{ yr}^{-1}$ and average mass flux of $0.34\text{ g-CH}_4\text{ m}^{-2}\text{ yr}^{-1}$ are very small in comparison with those from both geologic seepage and Siberian thaw lakes. The impact on atmospheric methane concentration is negligible. No anomaly of local atmospheric methane concentration has been observed during the open water season at Syowa Station [*Aoki et al., 1992*].

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Table 1 Dissolved methane concentration (DM) of lake waters during the open water season (Dec. 2003~Jan. 2004). 14 underlined data of DM at/near surface were used for flux estimation. ND means not detectable level <15 n mol L⁻¹. DO is dissolved oxygen concentration.

Lake (Type ^a)	Latitude ^b S	Longitude ^b E	Max.	Sampling depth m	DM depth m	Salinity nmol L ⁻¹	t _s ^c psu	DO °C	mg L ⁻¹
LANGHOVDE Area									
Yukidori Ike (2 or 3)	69°14.430'	39°45.352'		8.6	1	19.4	0	2.8	9.02
						66.6	0		9.54
Higashi-yukidori Ike (1)	69°14.243'	39°46.792'		19.5	1	20	0	2.5	9.45
					10	21.8	0		9.86
Tenno-kama Ike (1)	69°15.848'	39°46.196'		5	1	ND	0	1.2	10.1
					4	ND	0		10.2
Kami-kama Ike (2 or 3)	69°15.887'	39°45.871'		4.5	1	80.3	0	2.5	9.8
					4	19.3	0		9.8
Nurume Ike (4)	69°13.454'	39°39.400'		22.7	1	31.4	14.7	2.3	9.96
					12	233.6	31.8		10.26
Akebi Ike (3 or 4)	69°12.140'	39°39.088'		4.5	1	ND	4.8	2.9	9.97
					4	20	4.8		10.26
SKARVSNES Area									
Hotoke Ike (3)	69°28.591'	39°33.705'		3	3	209.2	1.3	1.4	12.87
Arisa Ike (4)	69°28.427'	39°37.368'		1.8	1.5	75.1	39.4	6.4	0.25
Oyako Ike (3)	69°28.526'	39°36.179'		8.5	3	384.5	0.2	3.5	15.6
Funazoko Ike (4)	69°26.821'	39°33.634'		7.6	6	19.7	202	9.0	
Suribachi Ike (4)	69°29.038'	39°39.671'		32	5	23.3	120	5.8	
					11	506.3	202		
					25	66.2	212		
Himebachi Ike (3)	69°28.900'	39°39.285'		2.7	1.5	51.8	0.2	3.0	10.23
Kobachi Ike (3 or 4)	69°28.989'	39°38.538'		12	1	114.4	7.9	0.9	11.59
					9	21322	16.5		11.92
Magohachi Ike (3)	69°29.277'	39°38.477'		5	1	27.4	0.6	2.2	9.95
					4	23.6	0.6		10.06
Namazu Ike (3)	69°29.997'	39°41.897'		21	1	29.9	0.6	3.5	9.41
					10	27.1	0.6		9.6
					15	27.8	0.6		
Ageha Ike (3 or 4)	69°26.739'	39°34.904'		5.5	4	31.4	3.5	1.9	10.49
Nezumi Ike (4)	69°26.805'	39°38.158'		3.1	1	258	6.8	7.4	9.84
					2	21	50		0.04

^a lake type

^b WGS 84

^c t_s: water temperature at the lake surface

Table 2 Deviations of k_{CH_4} estimated by various formulas ($t_s=2.7$ °C)

Reference	equation ($\times 10^{-6} \text{ m s}^{-1}$)	field	n	CH ₄ flux (g-CH ₄ yr ⁻¹)	deviation (%)
Nitingale [2000]	$k_{600} = 0.617 U_{10}^2 + 0.925 U_{10}$	ocean		0.5	1.269 0 (reference)
				2/3 (U<3.6)	1.253 -0.7%
Nitingale [2000]	$k_{600} = 0.64 U_{10}^2 + 0.28 U_{10}$	ocean		0.5	1.15 -8%
Wanninkhof [1992]	$k_{600} = 0.862 U^2$	ocean		0.5	1.5 19%
MacIntyer [1995]	$k_{600} = 1.25 U_{10}^{1.64}$	ocean		0.5	0.956 -33.7%
Cole [1998]	$k_{600} = 5.75 + 0.597 U_{10}^{1.7}$	lake		2/3 (U<3.6)	1.314 3.6%

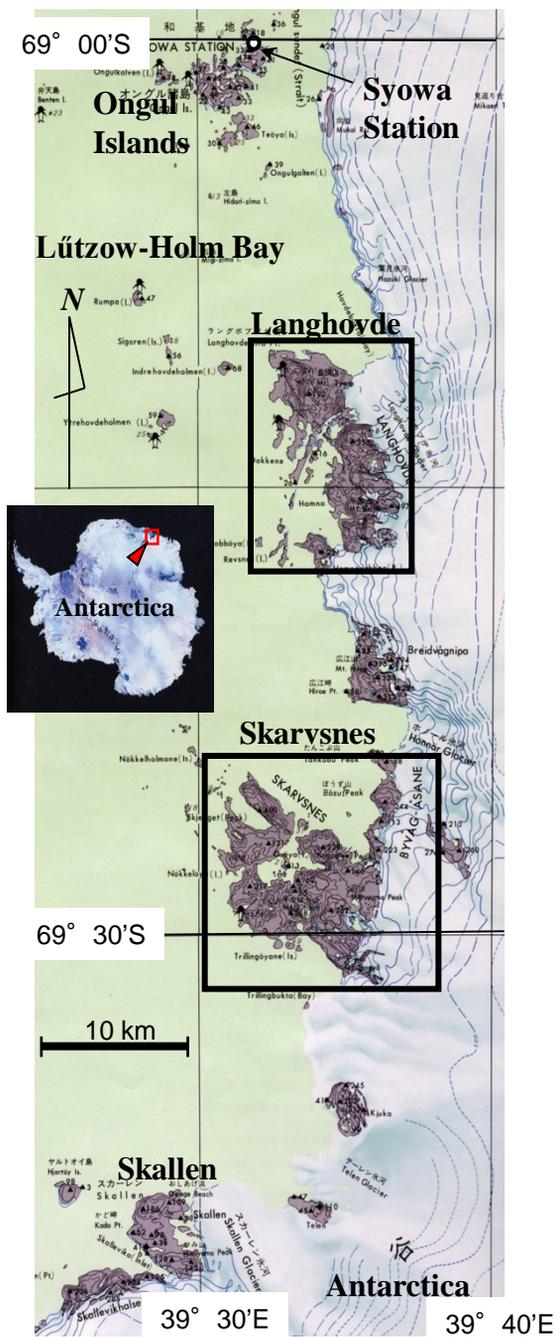


Fig. 1 Observation Sites: Syowa Oasis, East Antarctica (original map by Geographical survey Institute, Japan, 1989)

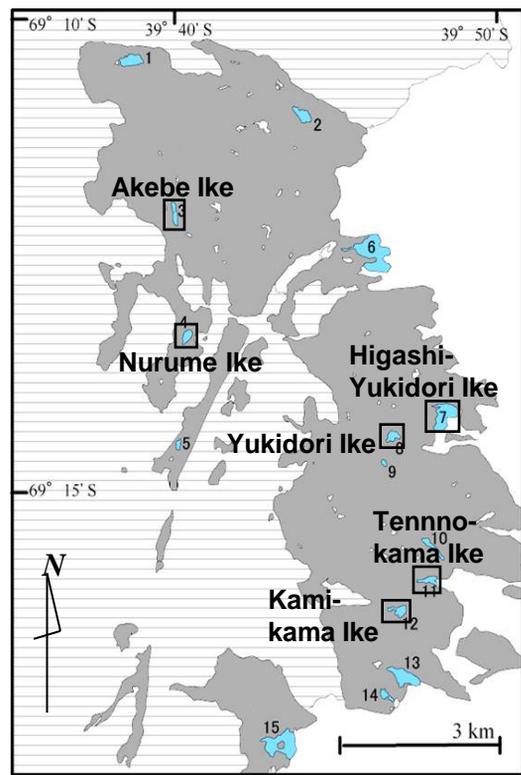


Fig. 2 Location of observed lakes (showed by boxes), Langhovde Area

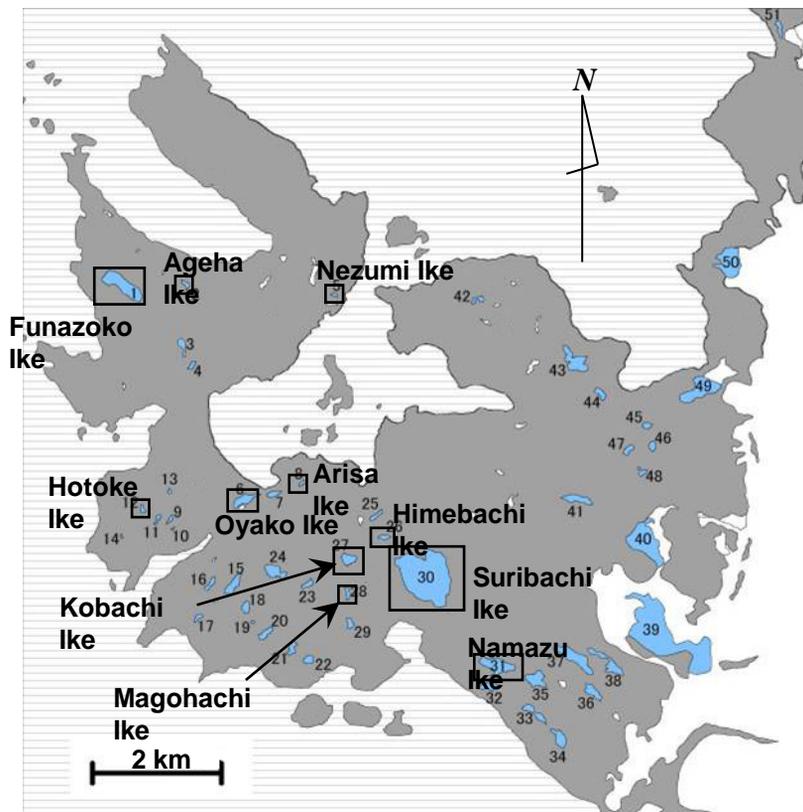


Fig. 3 Location of observed lakes (showed by boxes), Skarvsnes Area

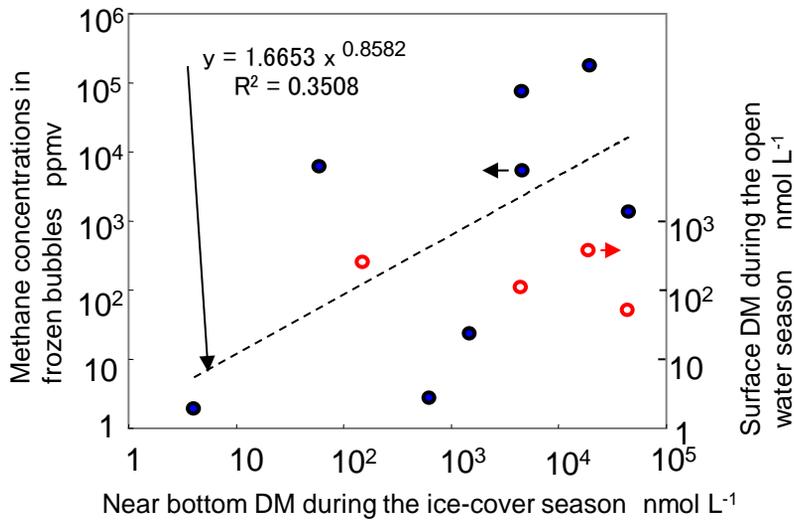


Fig. 4 Methane concentrations in frozen bubbles (solid circles) plotted against near bottom DM (DM_b : 0.5 m above bottom) during the ice cover season and surface DM (DM_s : hollow circles) during the open water season plotted against DM_b . The dotted line shows correlation of methane concentration and DM_b (Lakes in Skarvsnes area).

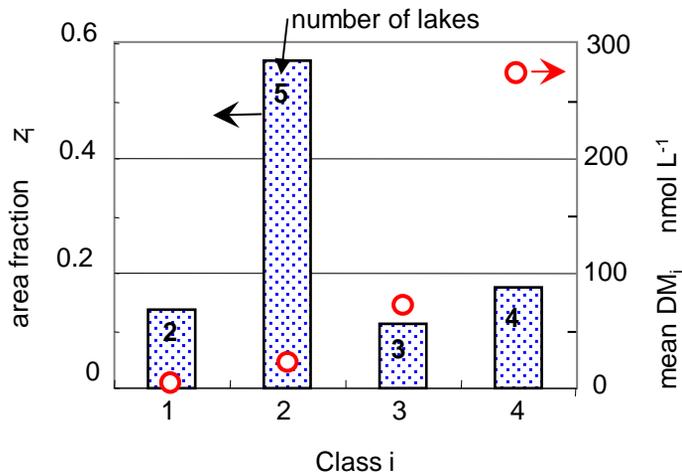


Fig. 5 Area fraction and areal weighted mean DM. Bar shows area fraction of each class z_i . Number in each bar is number of lakes in each class. Circle shows mean DM of each class.

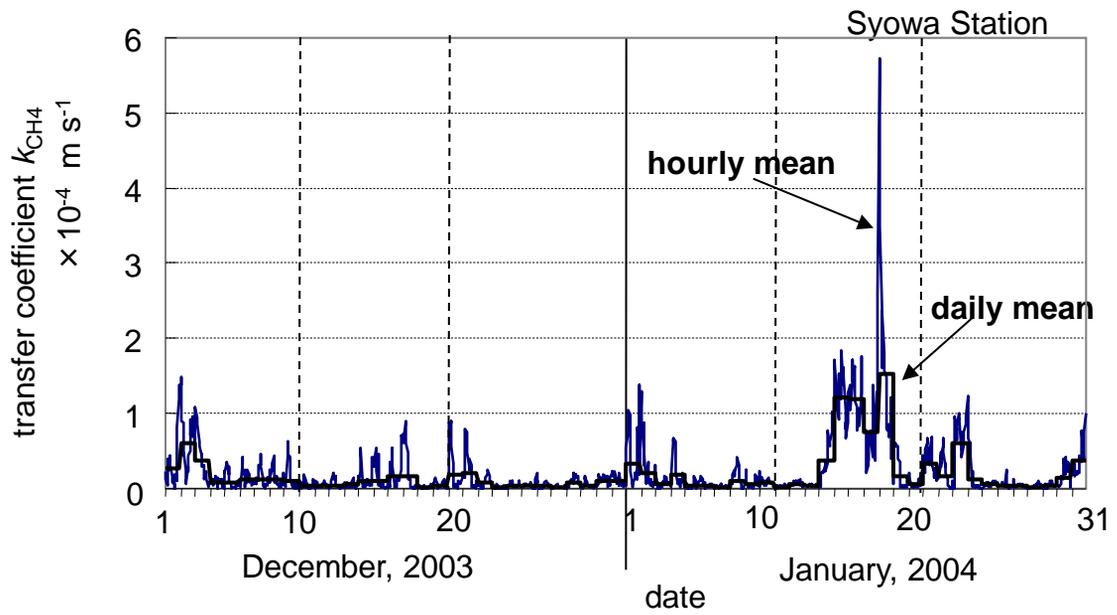


Fig. 6 k_{CH_4} changes with daily and hourly mean wind speeds using Nightingale's equation (9) for surface water temperature of 2.7° C