JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 114, D18114, doi:10.1029/2009JD011849, 2009

Methane efflux from bubbles suspended in ice-covered lakes in Syowa Oasis, East Antarctica

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Abstract

This is the first estimation of methane efflux from bubbles in lake ice in Antarctica. Bubbles suspended in shallow ice in 20 lakes were observed as 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 2004. Anomalous methane concentrations in bubbles suspended in lake ice and anomalous dissolved methane concentrations in lake water were frequently found. Methane concentrations in bubbles varied of five orders of magnitude, from 1.9 ppmv to 18% v/v. A procedure that makes estimations of methane flux from ice-bubbles possible has been developed, based on a relationship between bubble-density distribution, brightness observed by digital photographs and methane concentration in bubbles. Such a procedure applied to Lake Oyako Ike in the Skarvsnes area, where the maximum methane concentration was observed, suggests that total bubble volume is about 0.6% of ice volume and the mass of methane in bubbles in the lake is about 74 kg. Almost all gases in bubbles are released to the atmosphere in the early ice melt season (a period of a few weeks in December and January). By applying area fraction frequency distributions to methane concentration ranges for 20 lakes surveyed, extrapolation to the whole Syowa Oasis, including 110 lakes, would give a provisional estimate of total emission of about 2 tons-CH_4/yr.

1. Introduction

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). Since only vegetation of mosses and algae is usually found on the bottom of these freshwater lakes, although benthic copepods were found and they were first sampled at one of melomictic lakes (Lake Nurume Ike) in the Langhovde area in 2007 (Kudoh et al, 2008), it is thought that methane is produced by activities of anaerobic bacteria in the sediments of these lakes. Staff of the National Institute of Polar Research (NIPR) have 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). Imura et al (1999) found abundant colonies of mosses and algae, so called ‘moss pillars’. In order to find connections between biological activities and methane production, observations of dissolved methane concentrations (DM) were added to the project operations at JARE45.

Wand et al (2006) reported extremely high DM of 21.8 m mol/L 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. On the other hand, Andersen et al. (1998) found a relatively low DM level of 63-156 n mol/L 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 μ mol/L in the upper oxic waters, but increased below the oxycline to 1 m mol/L at an amictic, permanently ice-covered lake, Lake Fryxell, separated by Canada Glacier only 3 km from Lake Hoare. Bubble ebullition was not found using bubble traps for 40 days at the lake. These findings suggest that DM is strongly affected by various states such as geologic, microbial, meteorological states. On the other hand, any studies that reported methane concentrations in frozen bubbles in other areas in Antarctica could not be found.

The NIPR has also 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 CO_2, CH_4, CO and O_3 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. 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. The process is the same as air-sea exchange of gases (Liss et al., 1974; Nightingale et al., 2000). The other is direct efflux from bubbles suspended in lake ice early in the melting season. Zimov et al (1997) and Walter et al (2006) estimated ebullition of methane from thaw lakes in Siberia. They showed that thawing permafrost along lake margin were the most active methane source. On the other hand, however, there is no information on methane efflux from frozen bubbles at lakes in Antarctica.

The present study was undertaken to estimate the efflux of methane from frozen bubbles in seasonally ice-covered lakes. Observations of methane concentrations in frozen bubbles and DM in lake water were carried out by JARE45 in ice-covered season (April-November, 2004) in the Syowa Oasis, Antarctica.

2. Methods

2.1 Bubble and water sampling

Each location of water sampling in each lake was chosen at the point of maximum depth of the lake. A hole was drilled in the ice cover and water samples were obtained at the depth of 0.5 meters above the lake floor by using a Kitahara-sampler (1 liter). The water samples were sealed in 500 ml glass vials after an overflow procedure. Since DM analysis was completed within five days, no preservative was added.

Lake ice samples containing bubbles were cut with an oil-free chainsaw. The sampling locations, where the bubble densities appeared to be maximum, sometimes differed from the water sampling locations.

2.2 Measurements of methane concentrations in bubbles and DM

At Syowa Station, the ice samples were cut into small pieces using a hand saw. Each small ice piece containing bubbles was put into a 500 ml glass vessel (mouth diameter of 70 mm) completely filled with fresh water by a de-gasing procedure using a vacuum pump and an ultrasonic vibrator, and the glass vessel was sealed with a rubber membrane and a screw cap. The ice sample was melted at room temperature. Immediately after the gas in the bubbles had been completely released into the water (after about one hour), the free gas was extracted though the rubber membrane using a long needle attached to a syringe filled with pure water. The vessel was pressurized by the water to prevent contamination with room air. The syringe was sealed with a rubber cap after water purging and was placed in a water bath. Once a small hole on the bubble surface had been made by melting, bubble gas was released into the water within 10 seconds. Dissolution of gases in the water during this period was ignored. If the main component of the bubble gases was CO_2, the methane concentrations would change slightly because CO_2 dissolves more easily in water than do methane, nitrogen and other gases. The bubble 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. Nitrogen was used as the carrier gas. When the methane concentration of the first injection exceeded one hundred ppmv, the sample gas was diluted with a 500-10000-times larger volume of pure nitrogen. The errors of ±3% were added by the dilution procedure.

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 degrees C for more than 20 minutes, 1.625 ml of headspace gas was injected into the GC-FID. Standard gas (CH_4 of 1.98 ppmv balanced with N_2) filled into the same size vial with 10 ml of pure water by the de-gasing procedure mentioned above was used as a span gas. The minimum detectable DM by this technique was about 15 n mol/L. The analytical errors were within ±5%.

3. Site description and bubble formations

3.1 Study site and lakes

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, garnet gneiss, without organic sedimentary rocks. Raised beach deposits, which can be found up to 20 m above the 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 Skarvsnes area, as an example, are shown in Fig. 2. Almost all lakes (more than one hundred) have no ice cover for two months (no sunset season for 1.5 month from December to January) but are completely covered with ice for the other ten months. Since the depths of most of the lakes (2 - 32m) 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, vegetation of mosses and algae was still activated even during winter. The presence of moss pillars in Lake Hotoke Ike is impressive evidence of abundant vegetation. Syowa Oasis is windy area. Though the no ice cover season is the calmest, the maximum wind speed is about 40 m/s even in summer. Water column is usually mixed well by wind-induced convection in holomictic lakes.

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 not found around the coasts in Syowa Oasis (Sasaki, 2005).

The lakes in Syowa Oasis are divided into four different types (Seto et al, 2002). Lakes of Type 1 receive water and sediments from glacial melt. Salinities are generally less than 0.02 psu. Lakes of Type 2 (0.02-0.1 psu) receive water from glacial melt indirectly. Only water from snow melt comes into lakes of Type 3 (0.1-2 psu). Algal debris can be seen in both Type 2 and Type 3 lakes. Lakes of Type 4 have been isolated from the ocean by the uplift due to deglaciation in 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 dating ($AMS^{14}C$) of sediment core showed that lakes at an altitude of less than 8 m were isolated from the ocean later than 3500 years before present (yBP).

The thicknesses of sediments at the lake bottoms ranged from fifty centimeters to two meters. In some lakes, most of the sediment consisted of thick algal debris (sometimes more than one meter 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 for several lake cores (Matsumoto et al, 2006). High total organic carbons (TOC) were found in recent sediments that had formed after isolation from the ocean. The core top age was 300-400 yBP. It can therefore be assumed that methane is produced exclusively by recent microbial activity within lake sediments.

3.2 Bubble clusters and columns formation in lake ice

Many bubbles were frequently found in the lake ice in Syowa Oasis. Some of the bubbles were large (10-100 mm in diameter) as shown in Fig. 3 (a). Walter et al (2006) classified four distinct bubble clusters in thaw lakes in Northern Siberia. The bubble clusters observed in Showa Oasis are similar to the 'kotenok' in their categories. Arrays of bubbles (bubble columns) were also sometimes observed as shown in Fig. 3 (b). A mechanism of repetitive (not continuous) bubblings from the lake floor is necessary for the formation of vertical bubble columns. It seemed that these relatively large bubbles were formed at the ice-water interface by aggregation of small bubbles rising from the lake floor. Such a process can be easily reproduced in experiments in a cold room. If the bubbling continues to occur after maximum ice thickness is reached, gas simply pools beneath the ice. Such an example was found at Lake Kuwai Ike, Skarvsnes. A sampling hole (120 mm in diameter) drilled into the ice penetrated the gas pool. The gas blowout, shown in Fig. 4 (Matsuzawa, 2004), lasted for about ten minutes. Methane concentration was not measured, because the gas could not be sampled. The gas was not ignited by a lighter flame above the hole. It was not clear whether the gas was produced over a period of one year or over a period of many years.

Early in summer, solar radiation causes extensive sublimation, which leads to the release of methane to the atmosphere from near-surface bubbles. As ice-melt progresses, the number of bubbles that release gas into the air gradually increases. When the ice cover begins to melt, convections that occur in water columns near the open water areas cause transportation of higher DM to the surface and, as a result, methane gas is released to the atmosphere.

4. Results

4.1 Methane concentrations in frozen bubbles

The results of measurements of methane concentrations (mol fraction x_{CH4} in ppmv) in bubble gas and DM in lake water during the ice cover season in 2004 are shown in Table 1. The word “Ike” means “pond” in Japanese. The abbreviation FW and SW mean freshwater lake and saline lake, respectively. Saline lake was defined a lake with salinity of more than one psu. Measured salinities of the water sampled for DM measurements are shown in the table. The locations in Table 1 show the bubble sampling stations. Anomalous methane concentrations of more than 1000 ppmv (= 0.1% v/v) being found at one fourth of the lakes studied. Very high concentrations more than 10% v/v were found at three lakes (Lake O-Ike in West Ongul Island: 10.5% v/v, Lake Higashi-Ura Ike: 13.5% v/v, Lake Oyako Ike: 18.1% v/v). These levels in bubbles formed by microbial activity are not so anomalous in lakes in middle latitude. For example, the range of methane concentrations in bubbles (not frozen) at a pond above a sphagnum peat in Japan was 3-68% v/v (Sugimoto et al, 1997).

Methane concentrations varied in the order of 10^5 (1.9-180000 ppmv). Methane concentrations in many of the large bubbles (more than ten millimeters in diameter) were very high, indicating that the origin of the large bubbles was sediment of the lake floor. On the other hand, milky ice containing many small bubbles (around one millimeter in diameter) was also occasionally found, especially near the shores. Methane concentration in those bubbles was about the same as the atmospheric concentration, suggesting that the small bubbles were generated from dissolved air in the lake water. In these cases, the bubbles contained oxygen. Though O_2 could not be quantified in this study, the GC-FID had a little sensitivity for O_2 and a small peak could be seen at shorter retention time than that of methane. The abbreviation Y and N in Table 1 mean presence and absence of O_2 content in the bubble gases, respectively. Near atmospheric concentrations were often found in Type 1 and Type 2 lakes and very shallow lakes (< 2 m), and the gas contained O_2 in most of the cases. On the other hand, O_2 were not found in the bubbles that contained methane with high concentrations.

4.2 DMs in lake water

High DM anomalies were also frequently found near the bottom of the lakes in the ice cover season, e.g., DM of more than one micro mol per liter (=1000 n mol/L) was found at 60% of the lakes studied in the Skarvsnes area. A clear positive relationship between bubble methane concentration and DM was not found.

Both Lake Suribachi Ike and Nezumi Ike were hypersaline lakes, the maximum salinities of which exceed the scale of the instrument (40 psu). The salinity calculated from density near the lake floor at Lake Suribachi Ike was about 200 psu. Both are typical saline lakes of Type 4. Naganuma et al. (2005) reported a vertical distribution of salinity and halophilic bacteria in the water column of Lake Suribachi Ike and high water temperature (about 15 degrees C) near the lake floor. No large bubbles were observed in the ice, which was semi-opaque because of brine formation like sea ice, at Lake Suribachi Ike and Nezumi Ike. Extremely low DO and a strong smell of H_2S in water near the floor were always found at these meromictic lakes. Though DM anomalies were also observed in those meromictic lakes, the DMs at the surface were not always so high, perhaps because of strong stratifications. Such hypersaline lakes have also been found in other areas in Antarctica (van den Hoff et al., 1986; Vestfold Hills and Boswell et al., 1967; McMurdo Oasis, etc). Frenzmann et al (1991) reported extremely high DM of 4.9 m mol/L 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 μ mol/L) in the shallower layer. On the other hand, in Lake Suribachi Ike, DM was much higher near the boundary of stratification (11 m in depth) than that near the lake floor (30 m in depth).

5. Estimation of efflux of methane from lake bubbles to the atmosphere

5.1 Methane accumulation in frozen bubbles in Lake Oyako Ike

A full view of Lake Oyako Ike, which was taken from the hillside (height of 160 m, horizontal distance of about 500 m from the lake center) of Mt. Yokosima Yama (temporal name; 226 m in peak height), is shown in Fig. 5. Most of the snow cover has been removed by strong wind and sublimation just before summer (November), and the ice covers on the lakes can be clearly seen. Almost all gases in bubbles suspended in lake ice will be released directly to the atmosphere early in the ice melt season, though some of gas will be dissolved in lake water. Therefore, methane efflux can be estimated from the total accumulation of bubble gases and the methane concentrations. Estimation was carried out for Lake Oyako Ike in Skarvsnes as an example as follows. The ice surface was very smooth due to the polishing effect of sublimation. On the other hand, the top surfaces of a large bubbles suspended in ice were opaque and granular, while the bottom surfaces were usually smooth and transparent. Convection driven by the strong vertical temperature gradient of ice might occur in large bubbles. The bottom surface became very smooth by sublimation and the vapor rose up and was re-crystallized at the colder top surface. The top granular walls of bubbles reflect light well. Therefore, the mist-like (white) tone on the lake ice did not show the surface structures of ice but showed bubble clusters. If it is assumed that the size of all bubbles are similar, the bright and dark tone expresses the bubble number density.

Before analyzing, the rocks, snow covers and cracks were removed from the digital image to improve S/N of ice brightness data. Fig. 6 shows a histogram of brightness grade G (256 grades) of the analyzed area (shown by a box in Fig. 5). The vertical axis y (= pixel number at G / total pixel number) expresses the area fraction. The histogram showed that the brightness grade G for bubble clusters were ranged between 41-181 (grade lower than 41 indicating rocks and shadows and grade higher than 181 indicating snow covers and cracks). Calibrations were carried out at three stations in three different ways. At the first station, marked by □ in Fig. 5, no bubbles were observed (brightness grade G=$Gmin$ = 41). At the second station, marked by ○, bubbles were sampled and bubble shapes were observed, bubble sizes and methane concentrations were measured. At the third station, marked by △, a close-up picture, shown in Fig. 3 (b), was taken and the digital image was analyzed. The camera angle was calibrated by the ratio of short to long diameter in bubble images. The angle of the center axis to the horizon in Fig. 3 (b) was 25 degrees. Assuming the horizontal distribution of bubbles was a square grid, average horizontal density of bubble number was determined from Fig. 3 (b). Vertical distribution was corrected by the refraction coefficient of ice (= 1.31 at wavelength of 0.6 micrometers).

The correlation between brightness grade G and bubble number density D_N [bubbles/m^3] is shown in Fig. 7. An approximately linear relationship was obtained as shown in Fig. 7 and in Eq. 1 in the appendix below.

Average single bubble volume vb (= 0.60 Nml) was calculated assuming the bubble was rotational oval in shape with long and short diameters of 13.0 and 6.8 mm, respectively. These average dimensions of a bubble were determined from Fig. 3 (b). It was also assumed that the bubble volume is the same gas volume under normal conditions (0 degrees C, 1 atm). Total volume of bubbles was calculated to be 109 Nm^3, which corresponded to about 0.6% of ice volume. Average number density of bubble was 10,492 bubbles /m^3.

Methane concentration of bubble gas was about 18% v/v (mol fraction x_{CH4} = 0.18) as shown in Table 1. If it is assumed that all of bubbles in Lake Oyako Ike contain the same methane concentration, total methane volume can be directly obtained by the product of the methane mol fraction and bubble volume. As a result, total mass of bubble methane in the observed area (box in Fig. 5) was calculated to be 14.0 kg-CH_4. From production of the average bubble number density and lake ice volume, total mass of bubble methane in Lake Oyako Ike was calculated to be 74 kg-CH_4.

Annual average mass flux of methane efflux from lake bubbles is 1.37 g-CH_4/ m^2 (of lake area)/ yr. The annual average flux level is about a half of that from the Siberian arctic tundra (3.15 g/m^2; Wille et al., 2008).

5.2 Overall methane efflux from frozen bubbles in Syowa Oasis

Unfortunately, in the JARE45 operations, the required data set of bubble size, shape and distribution (number density), methane concentrations in ice core samples and clear ice surface photographs could not completely prepared except for at Lake Oyako Ike. Additional procedures were required to estimate methane effluxes from other lakes.

In visual observations of many lakes from an airplane, it seemed that the bubble number densities were similar with some exceptions. For estimating the methane discharge from all lakes in the Syowa Oasis area, it was therefore assumed that the average bubble number density is the same as that at Lake Oyako Ike analyzed above (and shown appendix below). On the other hand, methane concentrations varied in the order of 10^5 (1.9-180000 ppmv) as shown in Table 1. The results show that it is not appropriate to represent the methane concentrations for all lakes by the arithmetic average.

All of the twenty lakes studied except two hypersaline lakes were divided into six classes (from A to F) by methane concentration ranges. Average methane concentration x_i (i = A to F) for each class is shown in Fig. 8 (a). The histogram of lake area fraction z_i for each class can be obtained as shown in Fig. 8 (b). Corresponding lake number for each class is written in each bar in this figure. If it is assumed that the histogram in Fig. 8 (b) can be applied to all lakes in the Syowa Oasis area, total mass of methane in bubbles in this area can be calculated to be 2514 Nm^3. According to the results, the sum of methane in the lakes belonging to Class F (the highest concentration group), of which the area fraction was only 14.5%, corresponded to 77% of total methane accumulation in bubbles in Syowa Oasis.

When all of the methane has been released into the atmosphere early in summer (melting season), methane efflux from bubbles in the Syowa Oasis area is estimated to be about 1.8 tons per year. Annual average mass flux is 0.20 g-CH_4/ m^2 / yr. The flux was about one seventh of that from Lake Oyako Ike. This is the first quantitative information on methane efflux from frozen bubbles in Antarctic lakes, using average bubble size and average bubble number density, and applying a histogram of methane concentrations for twenty lakes to all (110) lakes.

6. Discussion

Etiope et al (2004, 2008) estimated that global geologic methane emission from deep seepage ranges between 43 and 64 Tg/yr (possibly approaching 80 Tg/yr). Emissions from individual seepage areas are in the orders of 102-103 g-CH_4/m^2/yr for mud volcanoes, and may reach orders of 105 g-CH_4/m^2/yr for large gas seeps, such as those producing "eternal flames" (Etiope, 2009). Microseepage, which is a diffuse exhalation of methane from soil, may range from a few units to thousands of mg /m^2/ day (Etiope 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 to 3.8 Tg-CH_4/yr for Northern Siberia, with an average mass flux of about 25 g-CH_4/m^2/yr. So, the estimated Syowa Oasis areal emission (0.2 g-CH_4/m^2/yr) is very small in comparison with that from both geologic seepage and Siberian thaw lakes. The impact on atmospheric methane concentration is negligible.

Since all bubble gases are released into the atmosphere within a few weeks (end of November – mid December), the flux during this period is about twenty-times larger than the annual average. No anomaly of atmospheric methane, however, has been observed at Syowa Station (Aoki et al., 1992).

There are large uncertainties in this procedure caused from several daring assumptions (applying a mean bubble size and density, a histogram of methane contents in bubbles to all lakes in Syowa Oasis, etc). Though the uncertainty of about ±50% may be exists in the estimation of methane efflux, it is important to clarify the order of magnitude of the flux in un-surveyed regions such as Antarctica. Further field observations are required in order to improve the uncertainty. Photographs of Lake Oyako Ike could be taken with good spatial resolution because the lake is located just beside Mt. Yokosima Yama. However, photographs of lake surfaces must usually be taken from an airplane or a helicopter. If an airborne SAR (synthetic aperture radar) can be applied to observations, the bubble number density can be determined even when the lake surfaces are covered with snow (Matsuoka et al., 1999). Walter et al (2008) tried estimation of ebullition from Alaskan lakes using SAR. Complete compositional analyses of bubble gas and isotopic analysis of CH_4 are however necessary in order to completely understand bubble formation process and methane origin.

The highest level of methane efflux from open water was observed during a brief period just after ice melt in high latitude lakes (Phelps et al., 1998). After ice cover melting in Antarctica, open water season lasts for about two months (December and January) and methane exchange between lake surface and atmosphere is dominant. This exchange during open water season should be estimated in order to determine total efflux of methane in Syowa Oasis.

Seasonally ice-covered amictic lakes, like the lakes in Syowa Oasis, are found at 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. Microbial activities and methane have generally been found, regardless of trophic status, in freshwater lakes at Signy Island, South Orkney Islands (Evans, 1982 and 1984). Lakes with no outflow in Schirmacher Oasis (Ramaiah, 1995) are also similar to lakes in Syowa Oasis. It seems probable that the procedure developed in this study can be applied to those lakes. Observations of methane efflux from ice bubbles and DM in lake water in various areas are expected in order to estimate total methane efflux from the whole Antarctic area.

Appendix A

An approximately linear relationship was obtained between brightness grade G and bubble number density D_N as follows (Fig. 7).

$$D_N = 136.94\, G - 5618.6. \qquad \text{(A1)}$$

In the histogram of G (Fig. 6), a small volume difference of ice dV at a small range difference of brightness grade dG can be written as

$$dV = H\, dA = y\, V\, dG = A\, H\, y\, dG. \qquad \text{(A2)}$$

The number of bubbles contained in dV is

$$dNb = D_N\, dV = A\, H\, y\, D_N\, dG. \qquad \text{(A3)}$$

The total number of bubbles in the observed area (ice volume) Nb can be expressed as

$$Nb = \int_{G\min}^{G\max} A\, H\, y\, D_N\, dG = A\, H \int_{G\min}^{G\max} y\, D_N\, dG\,. \qquad \text{(A4)}$$

The integration can be calculated as the sum of $y\, D_N$ for every G (dG = 1 grade) as follows:

$$Nb = A\, H \sum_{i=1}^{n} (y_j\, D_{Nj})\,. \qquad \text{(A5)}$$

where $n = Gmax - Gmin + 1$ ($Gmin$ =41, $Gmax$ =181) .

Total bubble volume Vb is

$$Vb = vb\, Nb = vb\, A\, H \sum_{i=1}^{n} (y_j\, D_{Nj})\,. \qquad \text{(A6)}$$

Average number density of bubble D_{Nm} is defined as

$$D_{Nm} = Nb\, /\, (A\, H\,) = \sum_{i=1}^{n} (y_j\, D_{Nj})\,. \qquad \text{(A7)}$$

Normal volume of total bubble methane in the Syowa Oasis Vb_S can be written as,

$$Vbs = vb\, D_{Nm}\, H\;\; As \sum_{i=1}^{6} (x_i\, z_i) \qquad \text{(A8)}$$

Notation

Variable

A lake area [m^2] (A of Lake Oyako Ike = 10157 m^2)

As total sum of all major lake (110 lakes) areas (= 8.92 km^2)

D_N bubble number density [bubbles/m^3]

d differential operator

G brightness grade (1-256)

H average maximum ice thickness (=1.7 m)

Nb number of bubble

V ice volume [m3]

vb average single bubble volume (= 0.60 Nml)

Vb total bubble volume

x_{CH4} mol fraction of methane = methane concentration

y pixel number at G / total pixel number

z area fraction = a lake area / total lake area

Suffix

b bubble

i class (A to F) of methane concentration

s Syowa Oasis (total)

Acknowledgements

The authors are deeply indebted to the members of the 45th Japanese Antarctic Research Expedition (JARE45) for assistance in cruising on sea ice and in samplings.

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Table 1

Methane mol fractions in bubble gas and DM concentrations of lake waters during the ice cover season in 2004

Lake	Latitude*	Longitude*	Saline/Fresh Salinity psu	Maximum depth m	bubbles** x_{CH4} ppmv	O$_2$	DM n mol / l
SKARVSNES Area							
Nise-Hyoutan Ike	69°29.223' S	39°37.637' E	FW	6	1.9	Y	–***
Hyoutan Ike	69°29.147'	39°36.835'	1.6	8	76285	N	4505
Kami-Tenpyo Ike	69°30.558'	39°43.489'	0.00	2	6073	N	60
Naka-Tenpyo Ike	69°30.319'	39°42.790'	0.01	3	23.9	Y	1469
Simo-Tenpyo Ike	69°30.130'	39°41.931'	8.7	2	2.7	Y	626
Himebachi Ike	69°29.223'	39°37.637'	2.1	2.7	1380	N	44784
Suribachi Ike	69°29.223'	39°37.637'	>40	32	No large bubble		1036 (11m)
			>40 (200)		–		187 (30m)
Kobachi Ike	69°29.223'	39°37.637'	17.6	12	5322	N	4598
Nezumi Ike	69°32.369'	39°15.866'	>40	2	No large bubble		153
Oyako Ike	69°32.046'	39°14.958'	0.7	8.5	180500	N	19163
LANGHOVDE Area							
Tenno-kama Ike	69°15.835'	39°46.609'	FW	5	2081	N	–
Kami-kama Ike	69°15.887'	39°46.196'	FW	4.5	2.9	Y	–
Higashi-Hamna Ike	69°16.952'	39°45.995'	FW	22.7	2.3	Y	–
SKALLEN Area							
Skallen O-Ike	69°40.656'	39°26.731'	FW	9.2	108	Y	–
West ONGUL Island							
O-Ike	69°01.340'	39°33.893'	0.3	10	105000	N	105
Ura Ike	69°01.531'	39°33.786'	FW	–	283	N	–
Higashi-ura Ike	69°01.476'	39°34.142'	FW	4.2	134700	N	–
East ONGUL Island							
Midori Ike	69°00.805'	39°35.189'	FW		368	N	–
Kamome Ike	69°01.067'	39°34.326'	FW	–	2.4	Y	–
Aragane Dam	69°00.352'	39°34.353'	FW	–	2.9	Y	–
Daiichi Dam	69°00.416'	39°34.803'	FW	–	2.8	Y	–
Mizukumizawa	69°00.447'	39°34.531'	FW	–	42.9	N	–

*WGS 84

**Bubble gas was diluted with nitrogen when the concentration was over 100 ppmv.

Y/N for O$_2$ shows presence/absence of small peak of oxygen in GC-FID chart.

***Mark ’ –‘ shows no data.

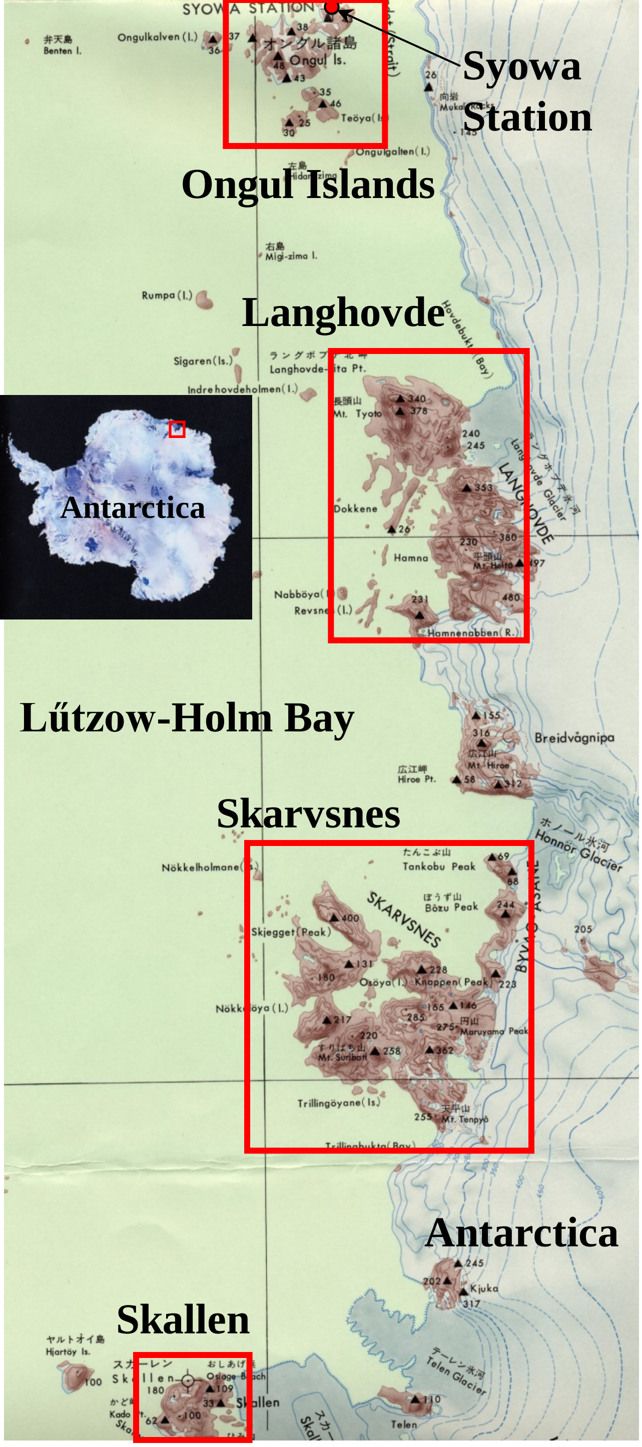


Fig.1 Observation Sites

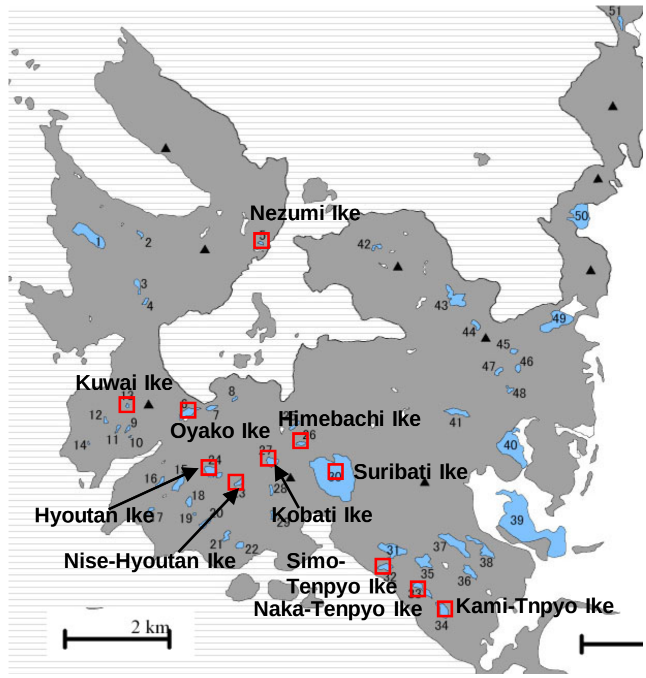
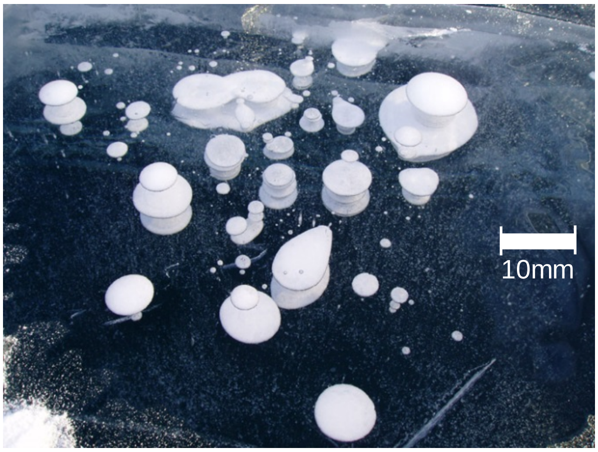


Fig. 2 Location of observed lakes, in the Skarvsnes area (T-) shows lake type (not authorized by the NIPR commission).



(a) Bubble cluster with big size bubbles in Lake O-Ike, West Ongul Island.
The scale is available on the picture center only. (Methane concentration of 10 % v/v)

(b) Bubble columns (Fig. 6 Point-a) in Lake Oyako Ike, Skarvsnes
Camera angle was 25 degree to horizon. The scale is available on the picture center only.
Average bubble size and average vertical and horizontal spaces of bubbles were determined from this picture. Methane concentration in the bubble gas was 18 % v/v.

Fig. 3 Bubble clusters that have high methane contents.

Fig. 4 Blowout of accumulated gas from a sampling hole (200 mm in diameter); Lake Kuwai Ike, Skarvsnes, 2004

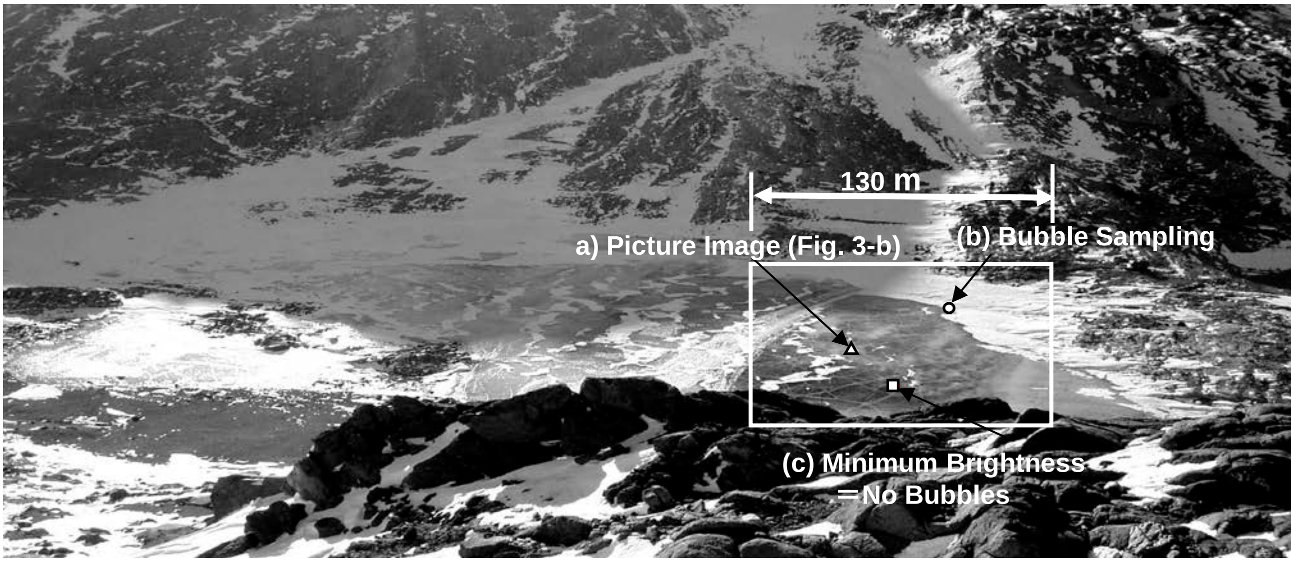


Fig. 5 Picture of Oyako Ike from a hill (Mt. Yokoshima) , Nov., 2004
White box shows the analyzed area with three calibration points;
(a) △ By bubble picture (Fig. 3-b),
(b) ○ By bubble sampling,
(c) □ Minimum brightness (No bubbles)

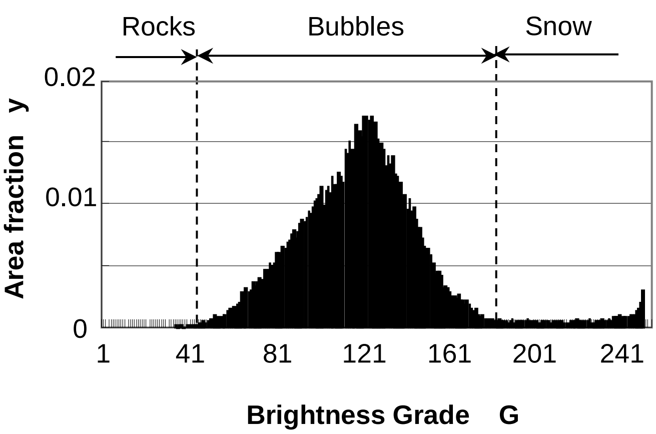


Fig. 6 Histogram of brightness level (256 grades) after removal of the data of rocks, snow covers and cracks. Brightness of bubble array was included within the brightness level range of 41-181.

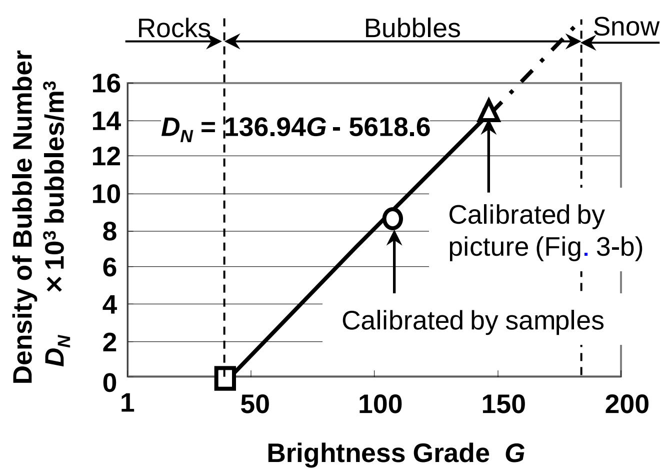
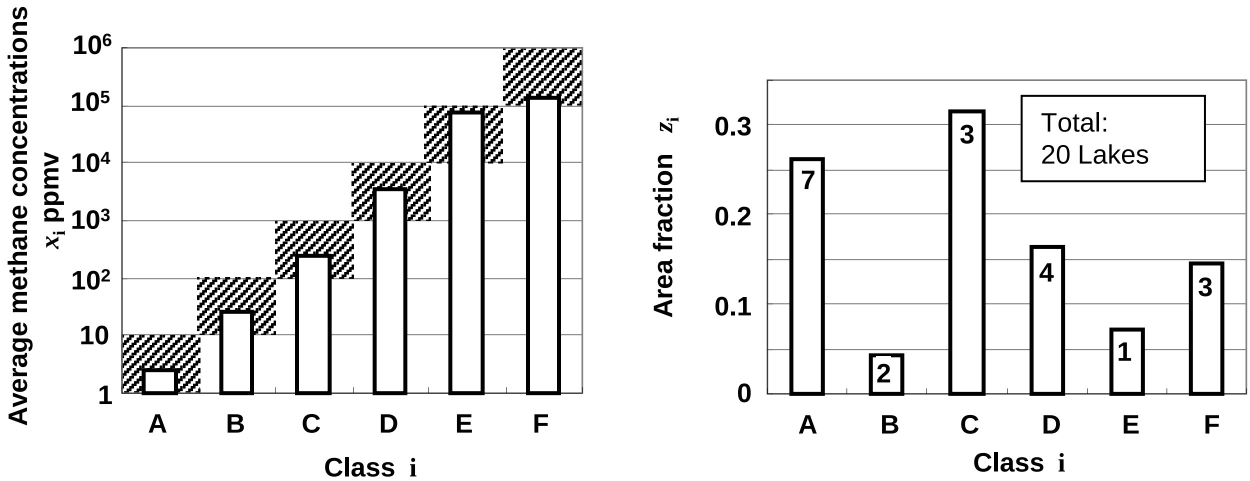


Fig. 7 Relationship between density of bubble number and brightness level.



(a) Classification of lakes and average methane concentrations in each class

(b) Area fraction of each class; Corresponding number of lakes is written in each bar.

Fig. 8 Six classified lake groups by methane concentrations in bubbles
Class A:1-10ppm, B:10-10^2ppm, C:10^2-10^3ppm, D:10^3-10^4ppm, E:10^4-10^5ppm, F:10^5-10^6ppm