Journal of Water Resources and Ocean Science
2014; 3(6): 89-94
Published online December 31, 2014 (http://www.sciencepublishinggroup.com/j/wros)
doi: 10.11648/j.wros.20140306.14
ISSN: 2328-7969 (Print); ISSN: 2328-7993 (Online)

Exchanges of methane between lakes and the atmosphere in Hokkaido, Subarctic Climate Region, Japan

Masafumi Sasaki, Noboru Endoh

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

Email address:
sasaki-m@mail.kitami-it.ac.jp (M. Sasaki)

To cite this article:
Masafumi Sasaki, Noboru Endoh. Exchanges of Methane between Lakes and the Atmosphere in Hokkaido, Subarctic Climate Region, Japan. *Journal of Water Resources and Ocean Science.* Vol. 3, No. 6, 2014, pp. 89-94. doi: 10.11648/j.wros.20140306.14

Abstract: Dissolved methane concentrations (*DM*) at twelve major lakes in Hokkaido, the northernmost island in Japan were observed in the open water season during 2006 ~ 2009 to estimate diffusive flux from lake surfaces to the atmosphere. An inverse relationship between lake size and *DM* was obtained in lakes in Hokkaido as was found for the European boreal lakes. All lake images larger than 0.001 km^2 were obtained by image processing of map data and area and number distributions were analyzed in order to calculate mass fluxes of diffusive methane. Total area of all (1,269) lakes in Hokkaido is 809 km^2. Regional diffusive flux of methane from lakes to the atmosphere in Hokkaido was estimated to be 0.581 Gg CH_4 yr^{-1}. Average diffusive flux density (per lake area) was about 0.718 g CH_4 m^{-2} yr^{-1}. This is a similar value to that in European boreal lakes on no-permafrost inland areas. Extremely high flux was found in Lake Abashiri-ko, one of highly eutrophic, meromictic lakes.

Keywords: Methane Flux, Lakes, Eutrophication, Global Warming

1. Introduction

Methane is well known as one of the green house gases (GHG). Lakes are significant source of methane to the atmosphere. In the previous work [1] [2], it was reported that the anomalous dissolved methane concentrations (*DM*) were frequently found even in Antarctica. In a subarctic climate zone, Hokkaido, the northernmost island in Japan, because of higher ambient and water temperatures and thicker organic sediments on the lake floors than those in Antarctica, it is expected that more active methanogenesis must be observed. If *DM* of a lake at the surface is supersaturated to the equilibrium for atmosphere, the lake acts as a methane source in the same manner of oceans [3].

In this study, field observations of *DM* were carried out mainly in twelve major lakes in Hokkaido in the open water season during 2006-2009. Diffusive mass fluxes of methane from all lakes in Hokkaido to the atmosphere are estimated from the results of the *DM* measurement and analysis of lake area and number distributions.

2. Methods

2.1. Site Descriptions

The locations of twelve observed lakes (circles) containing big-3 freshwater lakes in Hokkaido are illustrated in figure 1. The big-3 (70~80 km^2) freshwater lakes are Lake Kussyaro-ko, L. Shikotsu-ko and L. Toya-ko. The big-3 and L. Akan-ko are caldera lakes which have hot-sprig wells. Sampling stations are located on the shore of the lakes and off shore approached by a powerboat.

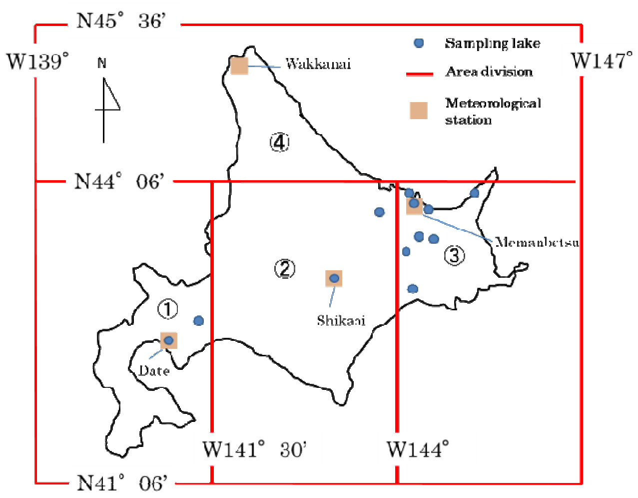

Fig. 1. *Observed Lakes in Hokkaido, Japan are shown by circles. Representative meteorological stations 1~4 are shown in solid square.*

2.2. Sampling Procedures

Surface water was sampled with a stainless steel pail and Van Dorn water sampler was used for sampling along water column. Temperatures and salinities of the water column were measured by a CTD. The waters were sealed in 500 ml glass vials after overflow procedure. Air was sampled in pre-vacuumed Tedler bags at the height of 1.5 - 2 meters from the ground.

2.3. Measurement of Dissolved Methane Concentrations (DM)

A headspace technique was applied to measure *DM* in water. A part of sample water in three glass vial (27 ml each) was replaced by pure nitrogen and 10 ml of sample water was remained in the vial. After each vial sample was heated and kept 60 °C for 20 minutes, headspace gas of 1.625 ml was injected to a gas chromatograph installed FID (GC-FID: Shimadzu GC-8A).

The measurements of DM were usually finished within 12 hours after sampling. When the samples had to be preserved for a few days or more, however, $HgCl_2$ was added as a preservative to the samples.

2.4. Measurements of Gas Concentrations in the Air

The sample air from a Tedler bag was directly injected to a 2 ml sample loop in the gas sampler which was installed in the same GC-FID mentioned above at the measurements of the methane concentrations in the air. Carbon dioxide concentrations were also measured through a methanizer located before FID. CO_2 escapes preferentially out of Tedler bag (3 % and 15 % of maximum reduction after one and three months respectively). The measurements of CH_4 and CO_2 concentrations were usually finished within 24 hours after sampling.

2.5. Estimation of Diffusion Flux of Methane from Lakes to the Atmosphere

A two-film model in air-water gas transfer was first proposed by *Liss and Slater* [4]. A great number of studies on air-sea exchange have been introduced and summarized by *Nightingale and Liss* [5]. Mass flux *m* (g CH_4 s^{-1}) is expressed as Eq. (1).

$$m = M_{CH4}\, k_{CH4}\, (DM - DM_0)\, A \qquad (1)$$

where, M is molecular weight, *k* is transfer coefficient, *DM* is dissolved methane concentration (DM_0 is equilibrium *DM* of atmosphere) and *A* is area of water surface.

The transfer coefficient of CO_2 in freshwater in the Schmidt number *Sc* of 600 at 20 °C, k_{600} was calculated using the empirical formulation Eq. (2) for air-lake exchange proposed by *Cole and Caraco* [6] .

$$k_{600} = 2.07 + 0.215\, U_{10}^{1.7}.\ (\mathrm{cm\ hr^{-1}}) \qquad (2)$$

The transfer coefficient of methane k_{CH4} can be obtained by the Schmidt number for methane, Sc_{CH4}. as Eq. (3)

$$k_{CH4} = k_{600}\, (Sc_{CH4} / 600)^{-n}\ . \qquad (3)$$

$$n = 0.5\ (U_{10} \geqq 3.6\ \mathrm{m/s})$$

$$n = 2/3\ (U_{10} < 3.6\ \mathrm{m/s})\ \text{by Deacon [7]}$$

$$Sc_{CH4} = \nu_{freshwater} / D_{DM}. \qquad (4)$$

Hourly mean wind speeds observed at four representative meteorological observatories were applied as U_{10} (equivalent wind speed at 10 m in height) to Eq. (2) during the open water season. These locations are shown in Fig. 1 (squares).

3. Results

3.1. DM Measurements

The results of measurements of *DM* in surface water are shown in Table 1. There are three big freshwater lakes (70~80 km^2), Lake Kussyaro-ko, L. Shikotsu-ko and L. Toya-ko in Hokkaido.

Table 1. *The results of measurements of DM in surface water in 2006~2009.*

Lake name	Latitude N	Longitude E	Area km^2	DM nmol L^{-1}
Kussharo	43°33.533’	144°20.350’	79.4	90
Shikotsu	42°46.300’	141°24.400’	78.4	34
Toya	42°34.993’	140°51.999’	70.7	27
Notoro	44°02.267’	144°10.388’	58.4	73
Abashiri	43°58.000’	144°10.000’	32.3	587
Mashu	43°34.694’	144°31.799’	19.2	41
Akan	43°27.000’	144°06.000’	13.0	114
Shikaribetsu	43°16.348’	143°07.133’	3.43	285
Mokoto	43°57.467’	144°19.933’	1.12	449
Rausu	44°01.826’	145°05.186’	0.43	501
Harutori	43°58.712’	144°24.361’	0.37	711
Tomisato	43°51.317’	143°45.200’	0.21	707

3.1.1. Lake Kussyaro-ko

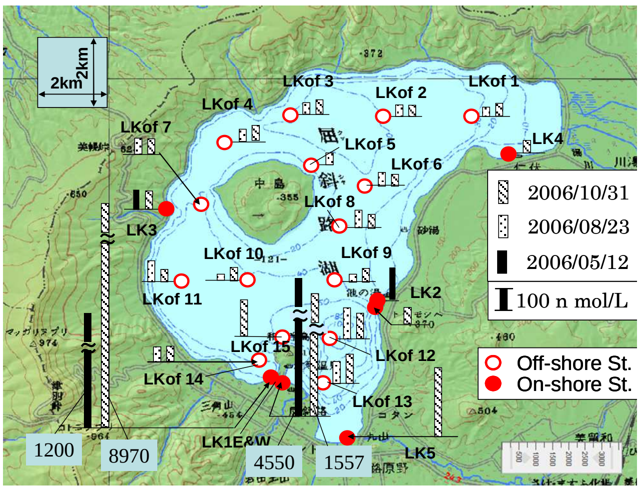

Fig. 2. *Surface DM distributions and seasonal changes in Lake Kussyaro-ko, 2006*

DM distributions at the surface and seasonal changes in Lake Kussyaro-ko in 2006 are shown in figure. 2. The surface *DM*s in northern area were lower than those in southern area, though the surface water is always super-saturated at every off / on shore station. Seasonal change of *DM* was not so clear in spite of change in water temperature through May to October.

Since L. Kussyaro-ko is an oligotrophic lake, lower methanogenesis activities at almost all stations are expected. However, there found extremely high *DM* (more than one μmol L^{-1}) along the shore of Wakoto Peninsula (St. LK1E and LK1W). Higher temperature than those at other stations suggested the existence of large amount of hot springs on the lake floor. One of the reasons of the *DM* anomaly may be higher activity of the methanogenesis because of such higher water temperature. Remarkable anomaly of methane concentrations in the air were seldom observed even around Wakoto Peninsula.

3.1.2. Lake Shikotsu-ko and Toya-ko

Both L. Shikotsu-ko and L.Toya-ko are typical oligotrophic lakes. *DM* distributions at the surface of L.Shikotu-ko on September 11 and that of L. Toya-ko on September 12 in 2007 are shown in figure 3 and figure 4, respectively. Both maximum *DM* are less than 50 nmol L^{-1} and *DM* are less than 30 nmol L^{-1} at almost all stations. It is considered that these *DM* distributions are typically shown in the oligotrophic lakes in Hokkaido, subarctic climate region.

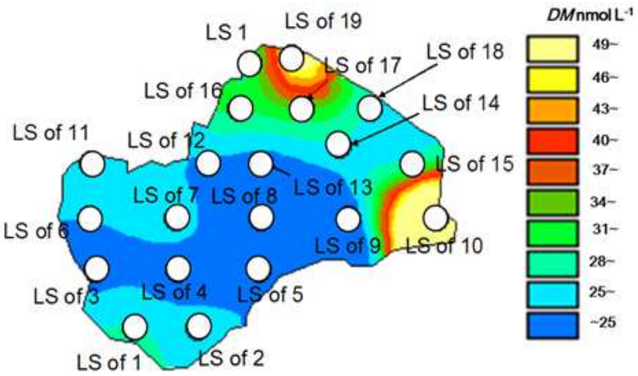

Fig. 3. Surface DM distributions in Lake Shikotsu-ko, on September 11, 2007

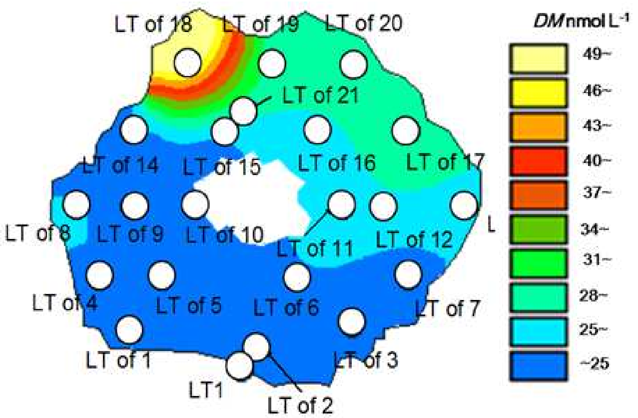

Fig. 4. Surface DM distributions in Lake Toya-ko, on September 12, 2007.

3.1.3. Lake Akan-ko

DM distributions at the surface and seasonal changes in 2006 in Lake Akan-ko, one of eutrophic lakes is shown in figure 5. Since seasonal changes in the surface *DM* can be clearly observed, it seems that the *DM* strongly depends on water temperature.

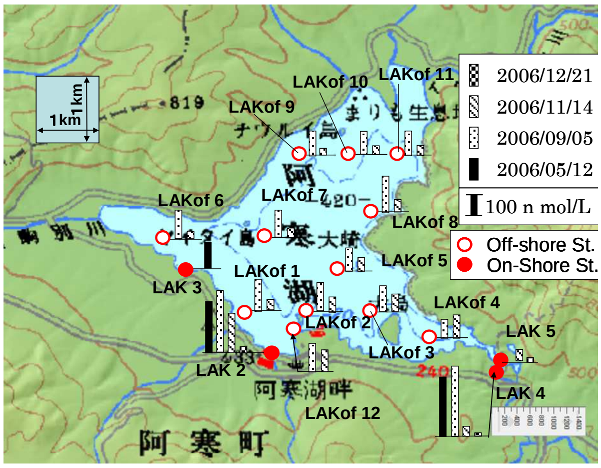

Fig. 5. Surface DM distributions and seasonal changes in Lake Akan-ko, 2006

3.1.4. Lake Abashiri-ko

Lake Abashiri-ko is highly eutrophic, meromictic lake. There is a clear saline layer at 5~7 meters deep. It has been eutrophicated by agriculture and pasturage along Abashiri inflow river. *DM* distributions at the surface in Lake Abashiri-ko are shown in figure 6. Super-saturated *DM* of 200 times or more than the equilibrium were found at all stations in summer. In early ice-covered season, *DM* under ice floe generally increased extremely (2~20 μmol L^{-1}) on-shore stations. Further study should be carried out to understand the relationship between these anomalous *DM* and the eutrophication or stratification.

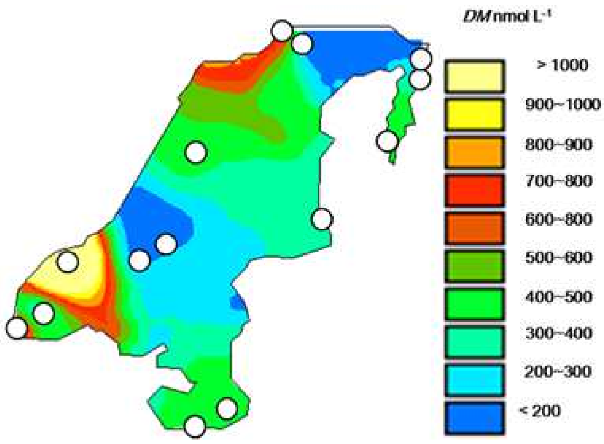

Fig. 6. Surface DM distributions in Lake Abashiri-ko on August 9, 2007

3.2. Relationship between DM and Lake Size

In order to determine average *DM* (=DM_{av}) in Table 1, a lake area was divided into pieces where each station located at near center as shown in figure 7 (Lake Kussyaro-ko as an example). The 'on shore' area was defined as the area of 0 ~ 10 m deep (painted out in figure 7) where the sun effectively lightened lake floor. Areal weighted mean DM can be defined as Eq. (5).

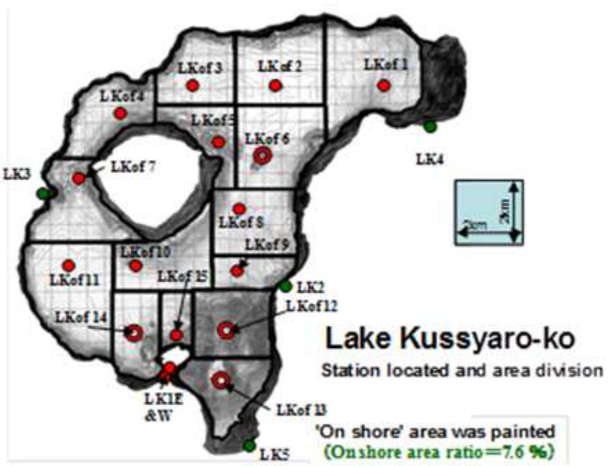

Fig. 7. Area divisions of Lake Kussyaro-ko (Circles show observation stations.)

$$DM_{av} = \sum_{K=1}^{Nst} (DM_K A_K / A) \qquad (5)$$

K: station number 1~$N_{station}$

Relationships between surface *DM* and lake area *A* are shown in Fig. 8. The dotted line in the figure shows Eq. (6) proposed by *Bastviken et al* [8]. As shown in Fig. 8, Eq. (6) does not express the *DM-A* relationship in Hokkaido, which has a steeper negative inclination as Eq. (7). One of the reasons of lower *DM* than those of Eq. (6) in beggar lake area zone is very low *DM* because of the big oligotrophic lakes as mentioned above. Another one of the reasons of higher *DM* than those of Eq. (7) in smaller lake area zone may be caused by higher water temperatures in Hokkaido than those in boreal lakes in Europe.

$$DM = 261.8\, A^{-0.227}. \qquad (6)$$

$$DM = 381.6\, A^{-0.498} \qquad (7)$$

(*DM* nmol L^{-1}, *A* km^2)

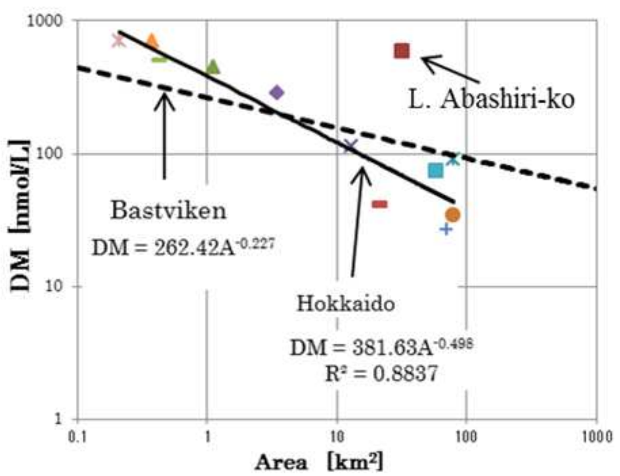

Fig. 8. Relationships between surface DM [nmol L^{-1}] and lake area A [km^2]

DM in L. Abashiri-ko becomes far from this *DM-A* trend (Eq. (7)) in Hokkaido as shown in Fig. 8. For example, *DM* in L. Mokoto-ko, which is also highly eutrophic, meromictic lake like L.Abashiri-ko, corresponds to the relationship Eq. (7). A unique production and migration processes of methane might be expected in this lake. Further observations of DM should be concentrated to L. Abashiri-ko in the future.

4. Estimations and Discussions of Methane Flux from Lakes to the Atmosphere in Hokkaido

4.1. Lake Size and Number Analysis (Individual Lake Area and Regional Histogram of Lake Area)

Map data (http://maps.google.co.jp/) were referred for lake area analysis. The map images were gradated into two images (black & white) using 'Adobe Photoshop'. Lakes were expressed as black images. The ratio of black pixels to total pixels was obtained from the histogram of each image (also made by Adobe Photoshop). The product of the black pixel ratio and the analyzing area shows the lake area. Images of rivers and seas were removed by hand before analysis.

All lake images larger than 0.001 km^2 were analyzed and area and number (A-N) distributions were obtained in order to calculate mass fluxes of diffusive methane using a commercial particle image analyzer 'A-zokun' made by Asahi Kasei Engineering Co. (http://www.asahi-kasei.co.jp/aec/business/sensing/product/azokun.html). Only scale conversion is required to apply this micron scale particles analyzer to kiro-meter scale lakes. The result of *A-N* analysis is shown in Table 2. Total area of all (1,269) lakes in Hokkaido is 809 km^2. Average lake to land area ratio is 1.03%, which is about 1/3 of global average (2.8% estimated by *Downing et al.* [9]). Class i (= 1~6) in Table 2 shows a lake size range between 0.001 ～ 1000 km^2. Representative value of *DM* for every class i was calculated using Eq. (7) for every average lake area.

Table 2. *Area and number (A-N) distributions of lakes from a map analysis in Hokkaido*

Class i	Lake area range		Number of lakes	Average area km^2	Total lake area km^2	Ratio of area to land
	Min km^2	Max km^2				
1	0.001	0.01	505	0.004	2.061	0.00003
2	0.01	0.1	544	0.033	18.121	0.00023
3	0.1	1	163	0.328	53.393	0.00068
4	1	10	46	3.256	149.754	0.00191
5	10	100	10	44.671	446.705	0.00570
6	100	1000	1	139.151	139.151	0.00177
Total			1269	0.638	809	0.01032

4.2. Estimations of Annual Methane Flux from Lakes in Hokkaido

Hourly mass flux Δm_{ij} at lake size class i was estimated by Eq. (8) (see Eq. 1~4) for a certain k_{CH4} calculated with an hourly mean wind speed U_{10} and monthly water temperature t_s at each representative meteorological station (j=1~4) as shown in Fig. 1.

$$\Delta m_{ij} = M_{CH4}\, k_{CH4ij}\, (DM_{ij} - DM_{0j})\, A_{ij} \times 3600. \quad (\text{g CH}_4\ \text{hr}^{-1}) \quad (8)$$

Annual mass flux m_{ij} (g CH_4 yr^{-1}) can be estimated by summation of hourly mass fluxes Δm_{ij} during the whole open water period. It is assumed that there was no air-lakes exchange during the ice-covered season. Total annual mass flux in Hokkaido is expressed as the total sum of m_{ij}.

$$m = \sum_{j=1}^{4} \sum_{i=1}^{6} m_{ij} \quad (\text{g CH}_4\ \text{yr}^{-1}) \quad (9)$$

Seasonal changes in mass flux of methane from lakes to the atmosphere in Hokkaido are shown in figure 9. Higher flux trend in summer is mainly caused by higher water temperature (= higher k_{CH4}) in this analysis.

Total annual diffusive methane flux in Hokkaido was estimated to be 0.581 Gg CH_4 yr^{-1}. Average flux density (per lake area) is 0.718 g CH_4 m^{-2} yr^{-1}. This is similar value to those in European boreal lakes on no-permafrost inland areas (0.923 for Swedish lakes by Bastiviken [8] and 0.784 for Finnish lakes by Juutinen [10]). And this average flux density is about five times greater than that from lakes in Syowa Oasis, in East Antarctica [2],

The annual methane flux in L. Abashiri-ko was estimated to be 0.066 Gg CH_4 yr^{-1}. The value corresponds to 11% of total annual flux in whole Hokkaido, while the lake area of Abashiri-ko occupies only 4%. The methane flux density in L. Abashiri-ko was 2.029. Such extremely high flux density suggests a unique mechanism of methane cycle. Further observations and analysis must be required for Lake Abashiri-ko.

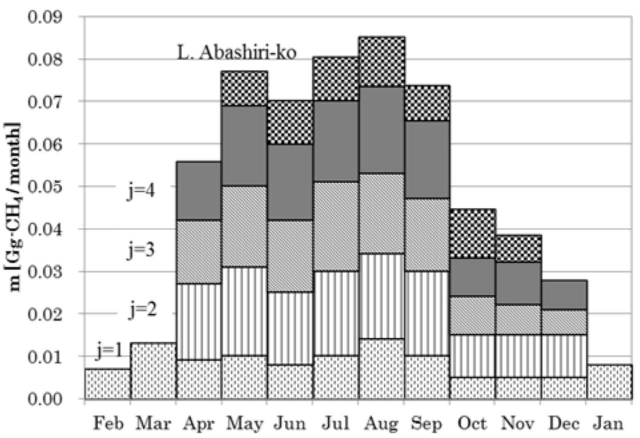

Fig. 9. *Seasonal changes in mass flux from lakes in Hokkaido.*

5. Conclusions

Observations of dissolved methane concentrations (*DM*) at twelve major lakes in Hokkaido, were carried out in the open water season during 2006 ~ 2009. Using the results and area histogram analysis of whole lakes larger than 0.001 km^2, diffusive methane flux from lake surfaces to the atmosphere were estimated. The study was concluded as follows,

1) An inverse relationship between lake size and DM was obtained in lakes in Hokkaido as was found for the European boreal lakes. The relation slope was steeper than that for the European lakes.
2) Total area of all (1,269) lakes larger than 0.001 km^2 in Hokkaido is 809 km^2. Regional diffusive flux of methane from lakes to the atmosphere in Hokkaido was estimated to be 0.581 Gg CH_4 yr^{-1}. Average diffusive flux density (per lake area) is 0.718 g CH_4 m^{-2} yr^{-1}, which is also comparable value for the European boreal lakes.
3) The annual methane flux in L. Abashiri-ko was estimated to be 11% of total annual flux in whole Hokkaido, while the lake area occupies only 4%. The methane flux density in L. Abashiri-ko was more than five times greater than that expected from the *DM-A* trend in Hokkaido. Such extremely high flux density suggests a unique mechanism of methane cycle. Further observations and analysis must be required for Lake Abashiri-ko to understand the carbon cycles.

Acknowledgements

We thank Shota Hirano, Yohei Baba, Toru Wakamatsu, Takuya Jin and Takahiro Kato for valuable assistances in the field works and the analysis. A part of this work was supported by the Japan Society for the Promotion of Science (#18651011).

References

[1] Sasaki, M., S. Imura, S. Kudoh, T. Yamanouchi, S. Morimoto and G. Hashida (2009), Methane efflux from bubbles suspended in ice-covered lakes in Syowa Oasis, East Antarctica., *J. Geophys. Res.*, 114, D18114, doi: 10.1029/2009JD011849.

[2] Sasaki, M., N. Endoh, S. Imura, S. Kudoh, T. Yamanouchi, S. Morimoto and G. Hashida (2010), Air-lake exchange of methane during the open water season in Syowa Oasis, East Antarctica., *J. Geophys. Res.*, 115, D16313, doi: 10.1029/2010JD013822.

[3] Liss P.S. and L. Merlivat (1986), Air-sea gas exchange rates: Introduction and synthesis, in The Role of Air-Sea Gas Exchange in Geochemical Cycling, edited by P. Buat-Menard , pp.113-127, D. Reidel, Norwell, Mass.

[4] Liss P.S. and Slater P. (1974), Flux of gases across the air-sea interface, *Nature* 247, pp.181-184.

[5] Nightingale P.D. and Liss P.S. (2003), Gases in seawater, in The Oceans and Marine Geochemistry, ed H. Elderfield, Vol 6 of A Treatise on Geochemistry (eds Holland and Turekian) Ch 3, 49 – 82.

[6] Cole J. J. and N. F. Caraco, 1998, Atmospheric exchange of carbon dioxide in a low-wind oligotrophic lake measured by the addition of SF_6, *Limnology and Oceanography*, 43(4), 647-656

[7] Deacon, E.L. (1977), Gas transfer to and across an air-water interface, *Tellus*, 29, 363-374.

[8] Bastviken, D., J. Cole, M. Pace and L. Tranvik (2004), Methane emissions from lakes: Dependence of lake characteristics, two regional assessments, and a global estimate, *Global Biogeochemical Cycles*, vol. 18, GB4009, doi: 10.1029/2004GB002238.

[9] Downing, J. A., Y. T. Prairie, J. J. Cole, C. M. Duarte, L. J. Tranvik, R. G. Striegl, W. H. McDowell, P. Kortelainen, N. F. Caraco, J. M. Melack and J. J. Middelburg (2006), The global abundance and size distribution of lakes, ponds, and impoundments, *Limnology and Oceanography*, 51(5), 2388-2397

[10] Juutinen, S., M. Rantakari, P. Kortelainen, J. T. Huttunenn, T. Larmala, J. Alm, J. Silvola and P. J. Martikainen (2009), Methane dynamics in different boreal lake types, *Biogeosciences*, 6, 209-223.