B-4.1 Field Measurement of Methane and its Flux from the Siberian Permafrost

Contact Person   Gen Inoue
                      Chief, Global Warming Mechanism Research Team
                      Division of Global Environment Research
                      National Institute for Environment Studies
                      2-16 Onogawa, Tsukuba, Ibaraki 305, Japan
                      Phone +81-298-51-6111 ext. 461 or 352,
                      Fax   +81-298-51-
                      E-mail inouegen@nies.go.jp

Abstract The estimation of methane emission from natural wetland is quite rough, and one of the main uncertainty is due to the luck of field measurement of methane flux in Siberia which has the largest wetland in the world. More important is the estimation of feed-back effect to the methane emission rate due to the climate change: Methane emission will increase as the result of spread of wetland due to the melting of permafrost originating from the outstanding temperature increase in high latitude.
  The methane flux has been measured on an observation airplane over the center of East Siberian Lowland as a joint research project with Central Aerological Observatory, Russia, in summer, 1992. The flux has been calculated by the aerodynamic method, 0.5-0.15 g/m2/day as an instantaneous value, and by the temperature inversion trap method to be 0.25-0.05 g/m2/day at night time as average value. This is an averaged flux over wetland, containing lakes, marshes, forests, etc., the flux from which are quite different depending on places. The averaged value seems to be more useful to estimate the total flux from natural wetland than the method to accumulate from individual ground-base measurement.

Key Wards   CH4, Global Warming, Siberia, Emission Rate, Wetland

1.Introduction
    Atmospheric methane has increased more than double in concentration since the industrial revolution, and it is currently increasing at a rate of 0.8 to 1.0% yr-1. Its source is widely scattered among many small ones. Some of them are anthropogenic source such as rice cultivation, enteric fermentation (mainly ruminants), landfills, biomass burning, animal wastes, coal mining, gas and oil drilling, etc. Natural sources are rather small in number; natural wetlands, termites, oceans, freshwater, and methane hydrate destabilization. The estimation of source strength has been summarized in the IPCC reports. The natural wetland is the largest source but the estimation is poor in comparison with other sources, especially anthropogenic sources. It is because the emission rate per unit area is rather small but the source area is wide, resulting a large source. The emission rate is very dependent on the conditions; water level, soil temperature, nutrient, and the history of them as well.
    The wetlands is usually divided into two groups; northern wetland, which are north of 40 N, and southern wetlands as the rest. The estimation of methane emission from northern wetland has several sources of uncertainties; i) Uncertainties in the area and the classification of the ecosystems, ii) uncertainties in the average flux attributed to the assigned ecosystems in a specific climate region, iii) uncertainties from seasonal and inter-annual variability in fluxes, and iv)

uncertainties due to measurement and analytical problems. The estimation of methane fluxes from Siberian wetlands is the main target of this series of research programs, and the final goal is to remove the above uncertainties.

The wetlands in permafrost area is expected to increase its area as the result of thawing permafrost in a warmer climate in future. The flux of methane in a different climate in future will be different from the present values. The fluxes may vary in a different way under a different scenario of Global Warming. These prediction will be reliable when and only when the comprehensive understanding on the mechanism and the parametrization of methane emission rate predicts the annual and inter-annual variation successfully.

In 1991, the research activities on this site have been reviewed, but it was found that there were no field research at all except two small groups: The methane contained in the ice wedge in permafrost at Tiksi has been reported by A.A.Arkhangelov[1], Moscow State University, and the methane flux measurement was under the work at Chersky by Gilichinsky, Institute of Soil Research and Photosynthesis.

2.Development of methodology of methane flux measurement

The most widely used method of methane flux measurement is so called "Chamber method", which is to measure the rate of methane concentration increase in a box covering the surface. The error of the rate measurement itself is within about 5%, but the flux itself is disturbed by the measurement environments as follows. The increase of air temperature in the chamber influence the flux seriously, and the measurement should be done before the temperature change caused by solar irradiation. The absence of wind is estimated to have little effect from the experiment with and without a fun in the chamber. The negative pressure in the chamber caused by sampling the inside air need to be escaped by compensation bag. The disturbance to the soil by putting a chamber on the ground is eliminated by the installation of a flame in about 20cm depth several days prior to the measurement where the chamber is put on it. The wooden street is constructed not to disturb by foot printing;The influence of a person walking at several meters distance is found to be serious because of fluffy character of peat. Only with these careful measurement environment, reliable flux data is obtained.

One of the method developed here is to estimate the flux in the night time by so called "Temperature inversion trap method". In West Siberian wetland, the wind velocity at night is small because of the little temperature difference in a homogeneous plain. Strong temperature inversion due to the radiative cooling of surface at night time is observed frequently. The methane emitted after the stabilization of atmosphere before the sunset is accumulated near the surface. In the morning next day, the inversion layer is destroyed as the ground is heated by solar radiation. The convex mixing starts from the atmosphere near the surface, and the height increases gradually. The concentration within this mixing layer is uniform because of good mixing at that time. If we measure the concentration at one altitude and the height of this mixing layer, we can calculated the total amount of methane which has been trapped in the temperature inversion since the sunset last day. The emission rate thus obtained is the averaged value both spatially and temporary. The difference of methane emission rate in day time and night time is not negligible, but not very serious because we can estimate this ratio from the data obtained by chamber method. The spatial averaging is rather preferable because the emission rate is very dependent on the places within several meters, and the averaged value is more useful to estimate the total flux from the wetland.

In order to estimate the total methane flux from a wetland, temperature inversion method is concluded to be the best method. This measurement can be improved if we use a balloon to obtain the vertical methane concentration profiles as we can continuously measure the

accumulation.

The method to calculate a flux from the gradient of concentration and eddy diffusivity is widely used and called as "Concentration Gradient method". For this purpose, the temperature difference at different heights and eddy diffusion coefficient are necessary to be measured. As the concentration difference measurement is a static one, conventional GC-FID analysis is enough. The eddy diffusivity is obtained by several method depending on the scale of it, and it was obtained by the vertical wind velocity measurement on the airplane here. Vertical wind velocity was obtained from the vertical acceleration and the attack angle of the airplane, both were obtained from INS signal, or an acceleration meter and an inclination meter. The accuracy is estimated about 15%.

3.Development of continuous methane measurement system on an aircraft

Gaschromatography with flame ionization detector (FID) is the standard method to determine the methane concentration in the atmosphere. However, the interval of this measurement is more than a few minutes and the distance between the samplings on the airplane is more than a few tens kilometers. The variation of the methane concentration over the wetland is in the scale of a few hundreds meters, and a lot of information is lost as the sampling interval is more than this scale. So it is essentially important to measure the methane concentration continuously.

In 1993, two continuous methane measurement system were examined on the airplane. One is to use FID without GC-column. In this case, the signal originating from none-methane hydrocarbons are added to the value of methane. In order to eliminate this effect, sample air is passed through Pt-Aluminum catalyzer in a temperature controlled furnace. If the temperature is properly set, larger hydrocarbons are oxidized and methane is not. As the oxidation efficiency is dependent on humidity, water vapor is removed completely by cooler at -60°C. The sensitivity is checked periodically by introducing two different standard gases, the base of which are air. The conventional GC-FID measurement has been done in parallel for calibration. Block diagram of the experimental setup is shown in Fig.1.
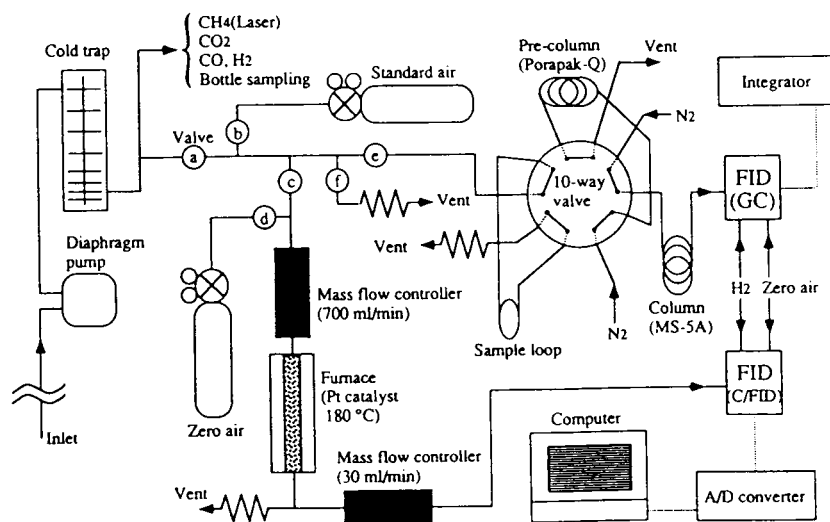

Fig. 1　Schematic of the CH$_4$ measurement system used in the flight experiment over Siberia. The CH$_4$ concentration is determined by the C/FID and gas chromatographic (GC) system.

The relation between GC and the catalyzer/FID (C/FID) signal obtained during the flight over West Siberia is shown in Fig.2. Two signal is in a good linear relation but the value from C/FID is higher than that of GC by 0.02ppm below 2ppm. The standard deviation below 2 ppm was

0.006ppmv. The C/FID value is higher at high methane concentration range which should be attributed to the contribution of non-methane hydrocarbon which is rich in concentration during the low altitude flight, where both methane and NMHC concentrations are high.

Fig. 2
Relationship between the CH_4 concentrations measured by GC and by C/FID system.

$y = -0.163 + 1.10x$

CH_4 (ppmv, C/FID)

CH_4 (ppmv, GC)
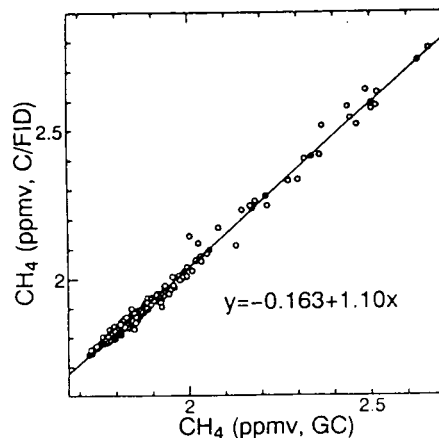
The other method is to use near infrared absorption measurement. As the stability of semi-conductor laser is excellent and the size is small in comparison with mid-infrared laser operating at liquid nitrogen temperature, we tried to use semi-conductor laser for methane concentration measurement. The small absorption coefficient at 1.6 μ m over band is compensated by a long pass cell, which is specially developed for aircraft measurement. This four mirrors system is newly designed to be perfectly stable against vibration as the change of angle of mirror is designed to cancel. Using the multi-dielectric coated mirrors fitted to this wavelength, sufficient light intensity was obtained after 200 times reflection (200m path length). Fig.3. Unfortunately because of trouble of laser, the measurement on board was not successful, but it achieved the accuracy of 1% at 1 second response in the laboratory.
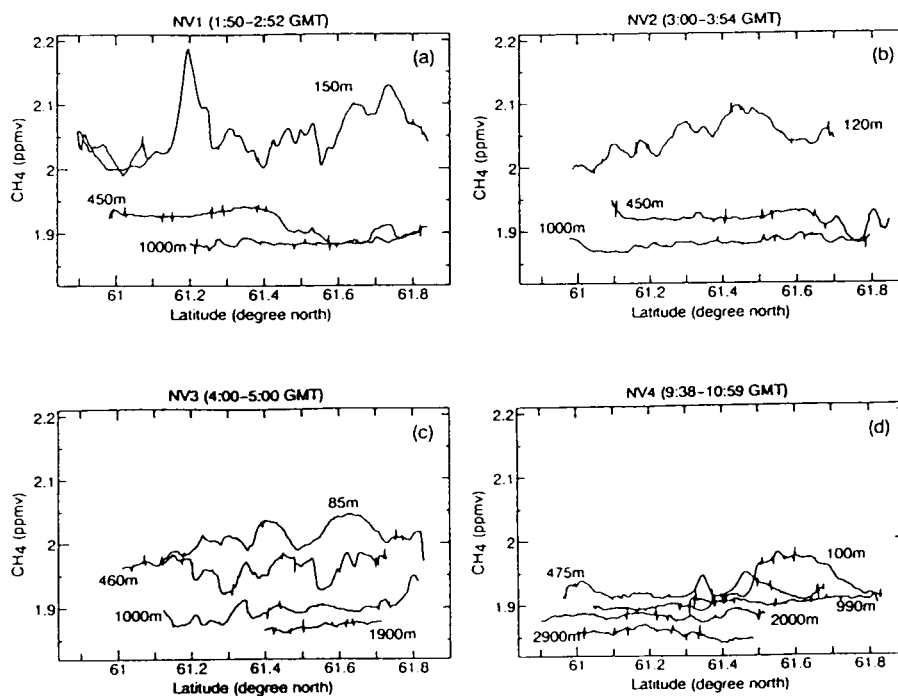
Fig. 3   Horizontal distribution of the CH_4 concentration observed in Nizhnevartovsk on July 21; (a) trial NV1, (b) trial NV2, (c) trial NV3, (d) trial NV4

4.Methane flux measurement on the airplane in West Siberia

Using a meteorological observation aircraft, IL-18, methane flux over the wetland in West Siberia has been performed in the periods of 18-22, July, in 1992 and 21-26, July in 1993. The aircraft belongs to Central Aerological Observatory (CAO), Federal Administration of Russian Federation for Hydrometeorology and Monitoring, and several navigation system such as INS, GPS and raider altimeter are equipped. A gaschromatograph with two FID, Hewlett-Packard model 5890 has been used with the continuous methane measurement system (Fig1) described above.

The typical flight pattern is as follows. The flight rout is fixed along the horizontal wind to minimize the effect of different ground level character. The wetland to the east of Nizhnevartovsk is practically uniform with a lot of shallow lakes and peat moss wetland. The flights at different altitude, say 150, 500 and 1000m above ground level, have been repeated to observe the dynamical change of the boundary layer.
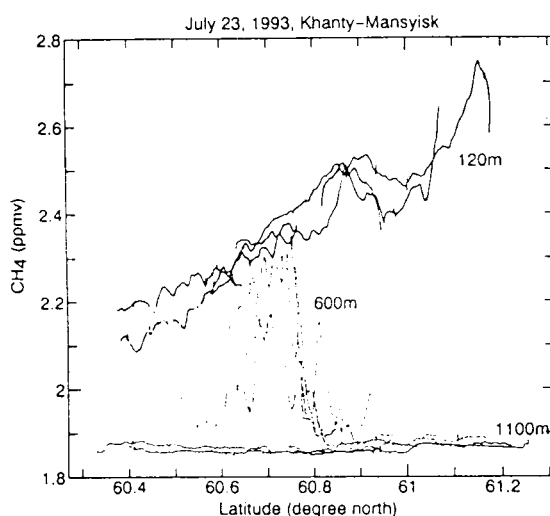

Fig. 4 Horizontal distributions of CH_4 concentration observed in Khanty-Mansyisk on July 23. Thick lines are the bottom distribution (120 m), gray lines are the middle distribution (600 m), and thin lines are the top distribution (1100 m) of CH_4

Fig.4a shows the horizontal distributions of CH4 concentration measured during 01:51-02:52 GMT, which is 07:51-08:52 in local time of Nizhnevartovsk. Fig.4b, c are the result of successive flight with about one hour intervals. Fig.4d is the flight 5.5hrs later. As the time passes, the methane concentration decreases at low altitudes and increases at high altitudes. These results show that the stagnant methane near the ground surface is diluted by overlying air in the troposphere. In this way the methane produced in the biosphere is transported to the free atmosphere. Horizontal distribution of the methane concentration at the lowest altitude of flight NV1 (Fig.-4a) shows large spatial fluctuations, which might reflect the spatial heterogeneity of the source strength and /or the different growth rate of the mixed layer. In Fig.5, the averaged methane concentrations of each horizontal distribution are plotted against the altitudes at different time. In the profiles of flights NV1 and NV2, which are similar to each other, large methane accumulation is observed at low altitudes. The profile of flight NV3, obtained one hour after flight NV2, shows that the methane concentration at the lowest altitude decreased but that at higher altitudes increased. This indicates that growth of the mixed layer occurred. The profile

of flight NV4, shows progressive vertical dilution of methane in the lower troposphere.

On July 23, 1993, the flight rout is designed to traverse two different area near Khanty-Mansyisk, South is water rich wetland and the north is relatively dry area. A lot of ponds are observed in the former region with little trees and main vegetation is peat moss. The latter area is covered by conifer forest mainly and wetland without trees are scattered in it (Fig.5). The methane concentrations are plotted with respect to the latitude here. When the aircraft repeatedly flew at the altitude of about 120m above the ground level, significantly enhanced methane concentrations were observed. The horizontal methane distributions at the altitude of 1100m were rather constant and as low as the background value, indicating that the top of the mixed layer did not reach the altitude of 1100m. The horizontal methane distributions at an altitude of 120m show that the concentration increases to the north. In contrast, the distributions at an altitude of 600m are higher in the southern part of the flight than that of north. Assuming that the concentration at 1100m is the value of background, the column amount of excess methane can be calculated for the north and south part of the flight divided at 60.8 degree north, giving 176 and 150 mg/m2 respectively. These values are similar each other although the concentration at 600m is quite different. This result suggests that the concentration difference between north and south is mainly due to the different altitude of mixing layer, i.e. the development of mixed layer. The mixed layer is better developed in the southern area than in the north, and it may be ascribed to difference in the insolation or the albedo of the ground surface, depending on the substances and the textures.

The vertical profiles of CH4 in the lower troposphere over the wetlands are summarized in Figure 5 together with those over two Arctic tundra area in Tiksi and Tazovskij. These profiles were taken at the first observation on each day. Vertical gradients of these profiles vary over a wide range. Throughout this study, enhancement of methane in the lower troposphere are significantly large in the wetlands, especially in Khanty-Mansyisk, while the concentration in the Arctic tundra region is almost the same as that in the unpolluted free troposphere. The vertical profile of CH4 concentration observed in the lower troposphere near the ground surface depends on the methane flux from the surface and the growth rate of the mixed layer. The rather small gradient of the profile at Plotnikovo, which locates the southern end of West Siberian wetland near Tomsk, is due to the strong vertical mixing because of late observation time of the day.
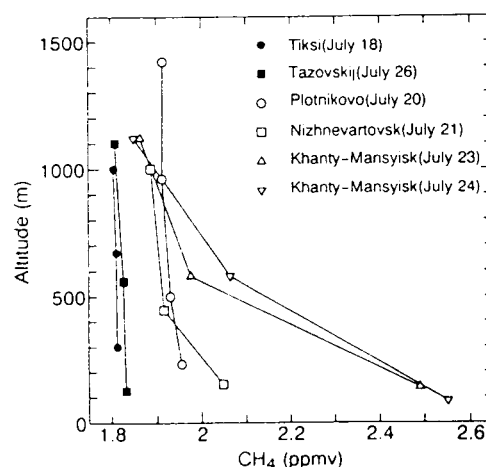

Fig. 5 Comparison of the vertical profiles of the CH₄ concentration obtained by intensive horizontal survey in the lower troposphere. Open marks indicate the profile over wetlands, and solid marks indicate that over tundra areas.

References

1) A.A.Arkhangelov and E.V.Novgorodova, "Genesis of Massive Ice at 'Ice Mountain', Yenesei River, Western Siberia, according to Results of Gas Analysis", Permafrost and Periglacial Processes, 12, 167-170(1991)