



The methane flux in the forest ecosystem : an automatic multi chamber method for continuous measurement

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MASTER FAGE
Biologie et Ecologie pour la Forêt, l'Agronomie et
l'Environnement

Spécialité
Fonctionnement et Gestion des Ecosystèmes

**The methane flux in the forest ecosystem:
an automatic multi chamber method for continuous
measurement**

SALLELES JADE

Mémoire de stage, soutenu à Nancy le 08/09/2010

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Structures d'accueil:



Forestry and Forest
Products Research
Institute - Kyoto,
Japan
&
Kyoto University



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1. Introduction

The countries that are signatories and mentioned in the B appendix in the Kyoto protocol have to reduce their greenhouse gases emission by 5% under the level measured in the nineties and methane (CH₄) is one of the most important greenhouse gases in the atmosphere, after the carbon dioxide (CO₂). It is less abundant in the atmosphere but it absorbs the infrared radiations 15 to 30 times more efficiently than the CO₂ (*Le Mer et al 2001*). Moreover, methane concentration in the atmosphere increased from about 700 to 1700 ppb within the two last centuries, especially from the industrial era, from 1750 (*Figure 1*).

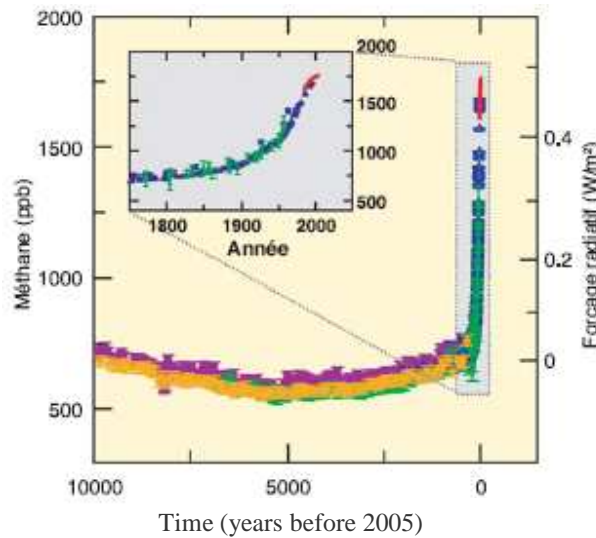
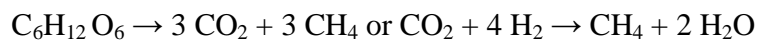
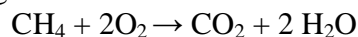


Figure 1 : Atmospheric methane concentration for the past 10,000 years and from 1750 (sidebar). Measurements are from ice carots (different colours corresponding to several studies) and from air samplings (red lines) (*GIEC 2007*).

60% of the sources of methane are anthropogenic (*Evans et al 2007*). They come from fuel exploration, rice production, biomass burning, waste management, wetlands... Whereas natural sources of methane are emitted by anaerobic bacteria called methanogenic bacteria through the reactions:



The methane emission from those bacteria occurs in anoxic conditions and depends on the level of the water in the soil and on the reductive conditions. The increase of the methane emission results to an imbalance between the sources and the sinks of methane. Major sinks for atmospheric methane are the reaction with hydroxyl radicals in the troposphere and in the stratosphere and the oxidation in terrestrial soils by methanotrophic bacteria (*Borken et al 2006*) through the following reaction:



Forests are mainly known to be a sink of methane but some peaks of methane emission have been found in particular places. Moreover, some soil disturbances can change a sink to a source of methane (*Dobbie et al 1996*). So, there are still strong uncertainties about the factors which influence the methane flux in the soil and in the forest ecosystem.

Until recently, most of the studies measured the methane flux between the soil and the atmosphere using manual sampling method such syringe or Tedlar bag methods. Those measurement campaigns allow only one measurement per day in the best cases. The large temporal variation in methane flux emphasizes the importance of methods that allow long-term and frequent monitoring of the flux, especially to estimate the annual emissions (*Akiyama et al 2009*). Continuous measurements would provide data for a long term period and assess the daily and seasonal pattern of the methane flux.

Los Gatos Research Company developed the Fast Methane Analyzer (FMA) which takes place in the new expertise of the forest ecosystem management. This analyzer measures the methane concentration continuously and at very low mixing ratios. The concentration may therefore be used to evaluate the methane flux from the soil. It is based on the absorption-spectroscopy method using near-infrared diode lasers. This tool is simple to use, rugged and compact, which makes it suitable for field studies.

The FMA arrived at the end of the year 2009 in the Kansai branch of the Forest and Forestry Products Research Institute (FFPRI) in Kyoto, Japan. I was in charge of its setting in the mixed Yamashiro experimental forest (YEF). The FFPRI focuses on the researches on the global warming and the utilization of forests and wood, to respond to the needs of the government and the society. The institute was established in 1905 in the south of Kyoto and was recognized as an incorporative administrative agency in 2001. During this work placement, I integrated the laboratory of Mr Kominami and Mr Miyama who work on the gas exchange in the mixed broadleaf species temperate forest.

The methane emission is measured for several years in Yamashiro site using a manual sampling method and a gas chromatograph. The measurements are focused on the emission by the stream in the forest. Indeed, a very high methane concentration was observed at some points. It attests that forests can sometimes be a source of methane and it confirms the need to evaluate the methane flux in the annual global flux estimation of this greenhouse gas. Purchasing the FMA will supplement the knowledge about the methane dynamic in the soil and will provide information about the temporal variations of the methane flux in the forest ecosystem.

The main objectives of this study were to set the FMA in the forest site and to calibrate it to measure the methane emission or the methane uptake in two different places continuously.

After a brief overview of the material employed in this study and the explanation about the FMA, this report presents the main results obtained by the FMA from May to July and tries to explain the variations of the methane flux from/to the soil to/from the atmosphere.

2. Material and methods

2.1. Site description

The first step began in a FFPRI room and consisted to measure the ambient air in order to understand how does the FMA work. The second step took place in the FFPRI garden in Kyoto, close to the building. The soil is occupied by deciduous and evergreen mixed broadleaf species. Finally, the third step took place in Yamashiro Experimental forest (YEF), in the south of Kyoto prefecture (34°47'N, 135°50'E), in a mountainous region of western Japan and located in a valley. It is composed of more than 50 deciduous broadleaf species (mainly *Quercus serrata*, 66% of the living tree biomass) and evergreen broadleaf species (mainly *Ilex pedunculosa*, 28% of the living tree biomass). The forest is a secondary forest that has been regenerated under a pine plantation. About 30 years ago, the pine wilt disease spread in this area and killed the *Pinus densiflora* (Jomura *et al* 2007). The forest canopy is closed and the height average about 12m. The site is one of headstreams of Kizu River.

The slope is gentle (about 5°) at the bottom of the valley where the measurement took place. The soil is underlain by weathered granite. It is thin, immature and sandy.

The mean annual temperature was 15.5°C, and the hourly minimum and maximum temperatures in 2002 were observed in August (34.8°C) and January (3.9°C). Annual mean precipitation from 1994 to 2002 was 1449mm.

2.2. Materials

The fast methane analyzer (FMA) relies on an off-axis Integrated Cavity Output Spectroscopy (ICOS). As the methane gas has a unique near-infrared absorption spectrum and because its wavelength is well-known it is possible to measure the gas concentration using the strength of its absorption. The technique consists in directing a continuous laser beam at a slight angle into a measurement cell defined by two highly reflectivity mirrors (Figure 2). The detector measures the fractional absorption of light at the methane resonant wavelength, which is an absolute measurement of the methane concentration in the cell (Hendriks *et al* 2008).

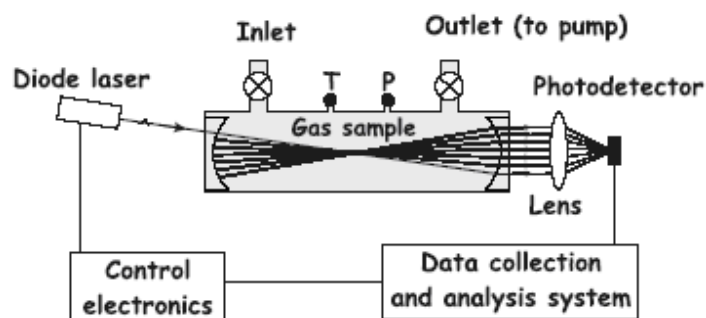
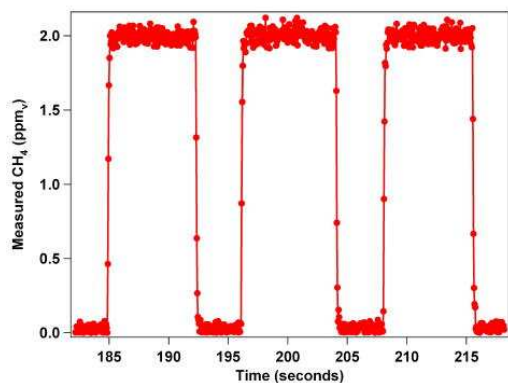


Figure 2 : Schematic diagram of the instrument based on off-axis ICOS for high-sensitivity absorption measurements in the near-infrared region. The measurement cell is comprised of a cylindrical tube sealed by a pair of high-reflectivity mirrors. “T” and “P” are the sensors of the temperature and the pressure of the cell (Baer *et al* 2002).



According to the technical data and the first tests conducted at the FFPRI, the response time is short (*Figure 3, Los Gatos Research 2009*) and the measurement accurate. Moreover, no external calibration is required because the methane concentration is an absolute measurement of the methane density in the cell.

Figure 3 : Response of the instrument to a repetitive square waveform over 30 seconds of N₂ flow (zero CH₄) and room air (2ppmv CH₄).

The operating temperature is wide (from 0 to 45°C). The FMA can be used in warm conditions like it occurs in YEF in summer. The storage capacity of the internal computer allows the internal hard drive to store the data almost indefinitely. The methane concentration (ppm), the pressure of the cell (Torr) and the temperature of the cell (°C) are recorded every second. A new data file is automatically created every 24 hours. The mirror values attest of the contamination of the cell but the mirror reflectivity is monitored and the measurement is compensated using the mirror ring down time.

Commonly, the methane concentration was measured by a gas chromatography (GC). In order to compare the data obtained with the FMA and the GC, a cross validation was performed with a GC equipped with a flame ionization detector (GC-FID) (Shimadzu (Kyoto, Japan) GC15A). The pack column of the GC-FID was a SHINCARBON ST (Shinwa Chemical Industry, Kyoto, Japan: SUS 2m x 3mm i.d.; column temperature 70°C; carrier gas N₂ at 60mL.min⁻¹; detector temperature 200°C; sample size 2mL). The calibration curve was determined using two standard gases (0.995 and 10.05ppm at 35°C, Sumitomo Seika Fine Chemicals Research Laboratory, Harima, Japan). The precision was 0.5% corresponding to the relative standard deviation based on 10 replicates measurements.

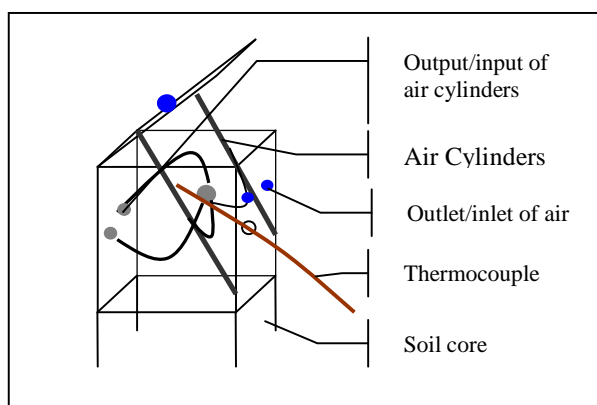


Figure 4 : Sketch of the automatic chamber.

The air was sampled in two automatic chambers made of polyvinyl chloride (PVC). Each chamber was based on a soil collar inserted of about 5cm into the soil. The total volume was about twelve litres (20x20x30cm). The chamber and the soil collar were sealed using a brown tape of 5cm width.

The lids of the chambers were opened and closed automatically by pneumatic cylinders controlled by an air compressor (*Figure 4*).



The inlet and the outlet of the air inside the chamber were set in a high position to avoid the mud entered the air sampling cycle (*Figure 4*). Both chambers were equipped with a switch in order to know which chamber was sampled and to select the data corresponding in the automatic flux calculation. The switch was fixed at the top of one side of the chamber using a strong double sided tape (*Figure 5*). An iron sheet fixed on the lid came into contact with it when the chamber was closed. A value less than 5V corresponded to a closed chamber (*Figure 6*).

Figure 5 : Switch set at the top of the chamber.

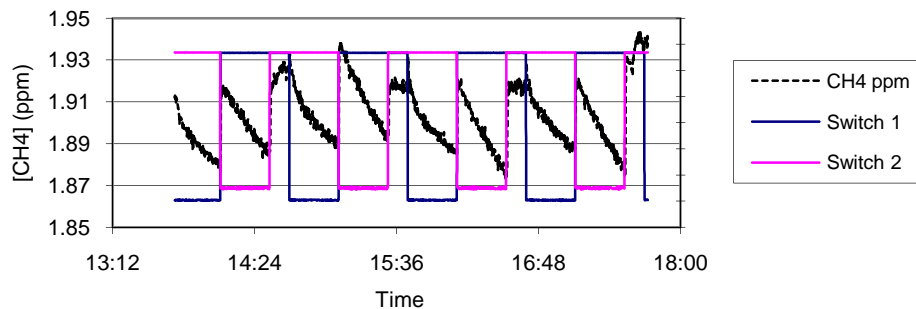


Figure 6 : Example of the methane concentration measurement in both chambers designating by the switches set at the top of the box, on the 13th of March.

In addition, the water vapour and the carbon dioxide concentrations were measured using an infrared gas analyser IRGA (LI-840, Li-COR, Inc., Lincoln, NE, USA). The air temperature in the chambers was measured with a thermocouple. The water temperature in the chamber 2 was measured with a thermometer. The soil water content was measured near the chamber at 5cm depth with a decagon device. All the data were recorded each second using a data logger (GL 200A midi Logger, Graphtec, Inc, Japan).

2.3. Method

The FMA arrived at FFPRI at the end of the year 2009. At first, only two automatic chambers were set. Afterwards, it is possible and planned to set more automatic chambers by increasing the connexions.



Figure 7 : The FMA

First of all, the FMA was switched on at the FFPRI laboratory, in order to measure the ambient air and to control the cell pressure and the cell temperature without any soil chamber. At this time, a cross validation was conducted. It consisted to analyze the air of 7 Tedlar bags filled with a standard gas at a known concentration (from 0.99 to 9.1ppm) by the FMA. Then, two automatic chambers were built and set in the forest soil in the FFPRI garden. The first measurement campaign began in the mid of February to the end of March. Finally, the FMA was carried in YEF on the 2nd of April. The first measurement begun on the 5th of April and

the first data were collected on the 6th of April. However, a leak was suspected in the first chamber, in the dry condition. The real data began from the 16th of April.

Both automatic chambers were set in the valley in YEF. The chamber 1 was inserted in dry conditions whereas the chamber 2 was set in wet conditions, in a stream of order 2 (Figure 8). The mean dry mass of leaf litter in the stream was about 1750g.m^{-2} (Miyama *et al* 2010). So, 70g of leaf litter was dried and set into the chamber 2 in order to reconstitute the natural conditions favourable to the methane emission.

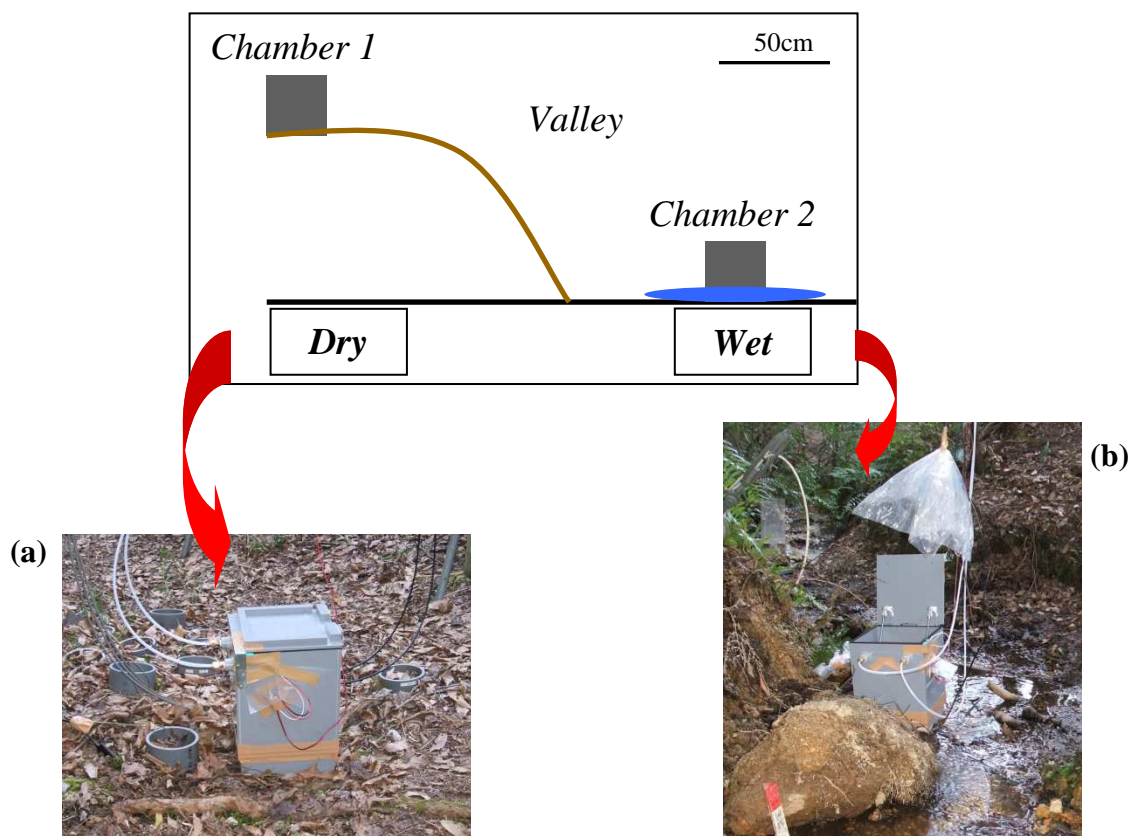


Figure 8 : Design of the experiment in YEF. (a) Chamber 1 in dry conditions, (b) Chamber 2 in wet conditions.

The FMA recorded the data continuously since it was on. The data were collected at least once a week.

Moreover, in summer, when the difference of the methane concentration during the sampling time in the chamber 2 was high, 6 air samples of 40mL were collected using a syringe. Those samples were analyzed by GC-FID at the laboratory in the evening following the sampling. They were collected at 15 minutes interval from the closure of the chamber for 1h15min. The gas sample was put into a 30mL glass vial topped with a butyl rubber stopper which has been evacuated beforehand at the laboratory. At the same time than the air sampling by the syringe, the methane concentration was analyzed by the FMA.

In order to observe the effect of the temperature on the methane emission, an experiment was conducted in the stream by adding two small heaters (used for aquariums, 18°C and 26°C) in the sediment under the chamber 2. They were switched on for about one week each one in its turn.

2.4. Automatic sampling system

The methane concentration was measured in a closed dynamic chamber. The system was composed of the FMA, the Li-COR device, 2 automatic chambers (20x20x30cm), tubes, pump, filters, two flow meters, 4 valves, 2 solenoid valves, a program controller (Zen 20C, OMRON) and an air compressor (*Figure 9*). A drier system was set after the LICOR device in order to remove the humidity of the air which arrives to the FMA. A Nafion tubing (Perma pure LLC) made of sulphuric acid removed the water vapour from the sampling line by the chemical affinity of the water compound to the acid. Once the water was absorbed into the wall of the Nafion tubing, the water permeated from one sulfonic group to another until it reached the outside wall of the tubing, where it evaporated into the air and trapped into a desiccant box.

The sampling time was 25 minutes for each chamber with a waiting time of 10 minutes while the air was sampling from the chamber 2. Therefore, there was one measurement per chamber per hour. This sampling time was a trade off between the time required to measure the methane concentration and the time required to let the chamber opened and aerated.

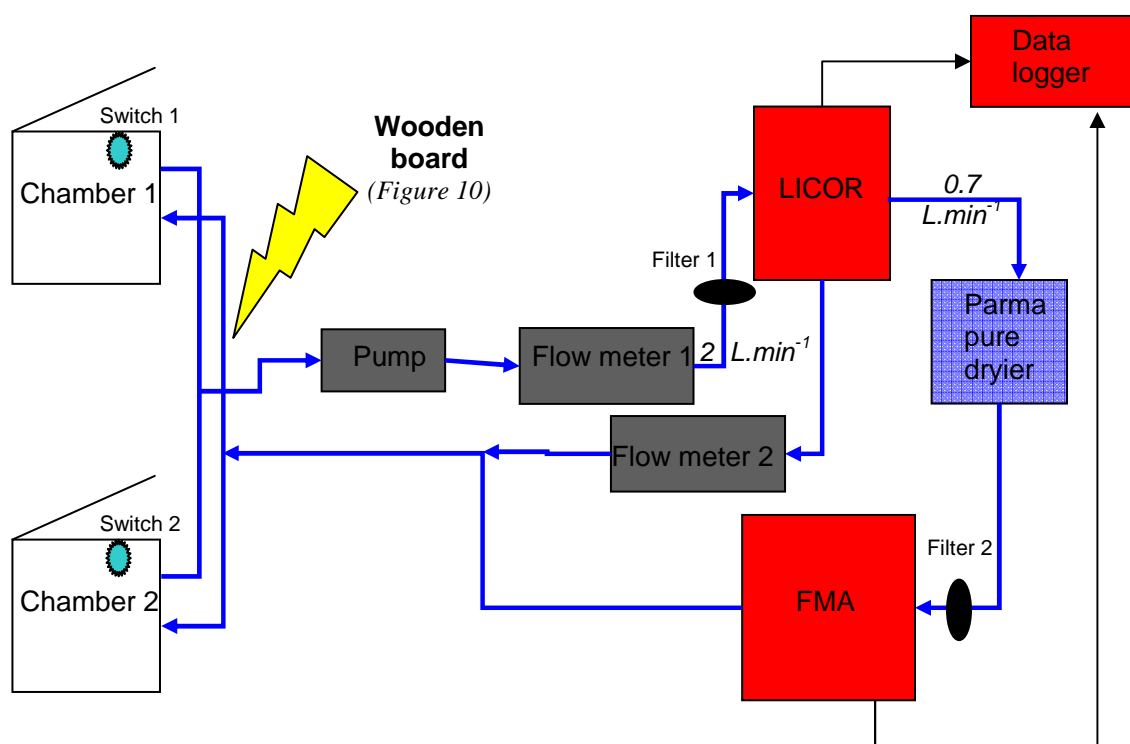


Figure 9 : Connexion of the two automatic chambers and circulation of the air in the closed system.

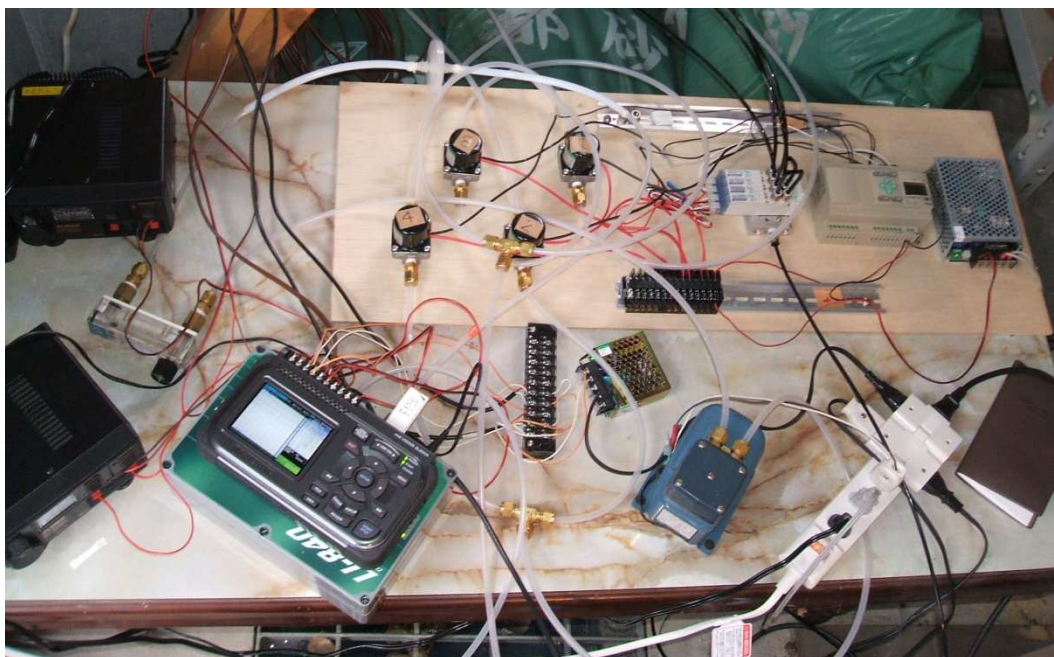


Figure 10 : The wooden board set in the FFPRI building.

2.5. Data analysis

The methane concentration through the time measured in the automatic chambers was assumed to be linear. So, linear regressions were performed on each continuous measurement to calculate the methane fluxes rates, using air temperature within the chamber headspace. It was executed by a Fortran program. The methane flux was calculated as follow:

$$F = \rho * \frac{V}{A} * \frac{dC}{dt} \quad \text{Équation 1}$$

where F is the methane flux ($\mu\text{g.m}^{-2}.\text{s}^{-1}$), ρ is the density of the methane in the chamber multiplied by the ratio of the total volume (V) to the soil area (A) and $\frac{dC}{dt}$ is the linear change in gas concentration in the chamber.

The correlation coefficient was calculated in order to test the linearity of the increase/decrease during the 25 minutes enclosure. All the negative methane fluxes (net absorption by soil) met the selected linearity criterion (95% confidence), giving correlation coefficients of 0.85-0.99. However, the positive methane fluxes (net emission by soil) did not meet this criterion from April to June because of the scattered data. Indeed, the methane emission in spring was low and the variability high. However, the criterion was reached in summer.

To calculate the gas flux, several quality controls were used: the sampling time (25 minutes), the first and last two hundred seconds were not taken into account to avoid the disturbance of the lid movement and the contamination by the previous measurement, the absolute value of the methane concentration (more than 0 ppm and less than 4ppm) and the r-square of the linear regression.

The software R (2.9.2) was used to perform the statistical correlations. Means and medians were calculated according to their confidence intervals at $P < 0.05$ (\pm CI). Graphs were made using Excel.

3. Results

3.1. Setting of the Fast Methane Analyzer

The data obtained when the fast methane analyzer was set near the FFPRI garden were an example of what it could be observed in the field site (*Figure 11*). The order of magnitude of the CH_4 and CO_2 concentrations, the flux calculation, the possible leaks, the movement of the automatic lids of the chamber and the scale of the Midi Logger device were checked.

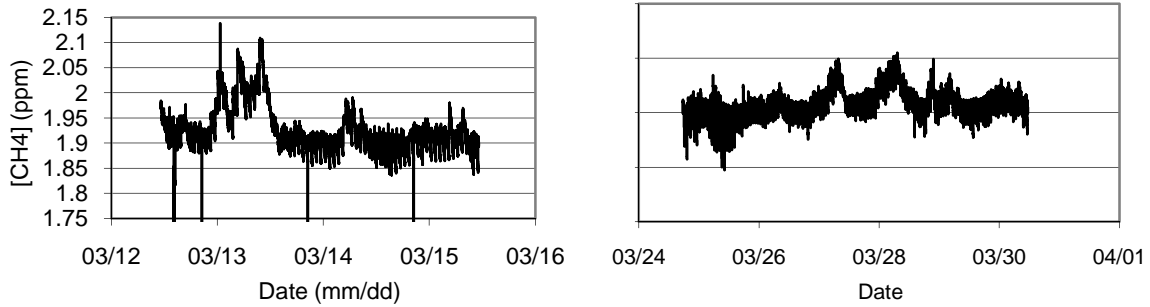


Figure 11 : Methane concentration in both chambers from the 12th to the 15th of March and from the 25th to the 30th of March at the FFPRI garden in March.

The strong peaks of the methane concentration (very low or very high) corresponded to the disturbance generating by the closure and the opening of the chamber by the air cylinders. Once the FMA was set in the field site, and after the leak checking, the data were collecting once a week from the GL 200 and the FMA, from the beginning of spring, in April, to the mid of July. The chamber 1, set in the dry conditions, showed a net methane absorption by the soil according to the decrease pattern of the methane concentration measured by the FMA. On the other hand, in the chamber 2 set in the river stream, an increase of the methane concentration referring to a net methane emission was registered (*Figure 12*).

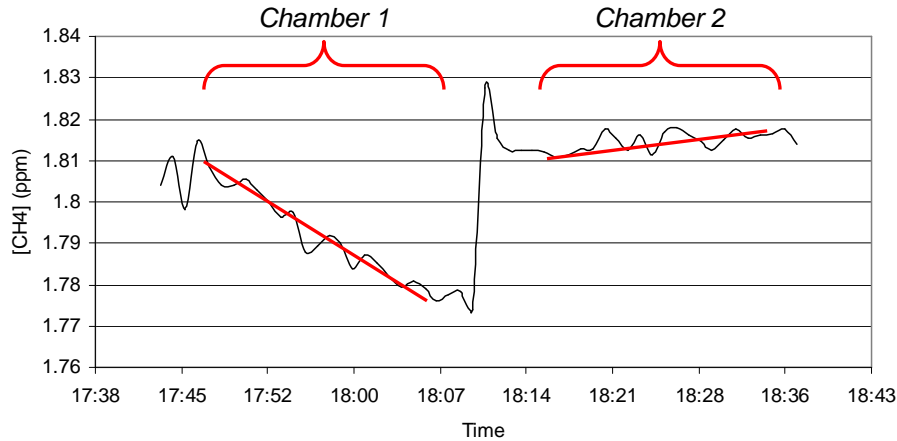


Figure 12 : Example of the trend of the methane consumption in the chamber 1 and the trend of the methane emission in the chamber 2, on the 15th of May. The 200 first data are cut in the flux calculation.

In spring, the methane concentration in the chamber 1 decreased of about 0.030ppm in 25 minutes (from 0.025ppm to 0.040ppm). It was twice as high as in the chamber 2 where the decrease was about 0.0015ppm. In summer, the difference of the CH_4 concentration during the sampling time in the chamber 1 was similar than in spring but it was between 0.034 and 4ppm in the chamber 2.

3.2. Cross validation

A clear relationship was found between the calibration gas and the methane concentration measured by the FMA in a dry air (*Figure 13, (a)*). In addition, the methane concentrations measured by the GC-FID and the FMA for about one hour were similar (*Figure 13, (b)*).

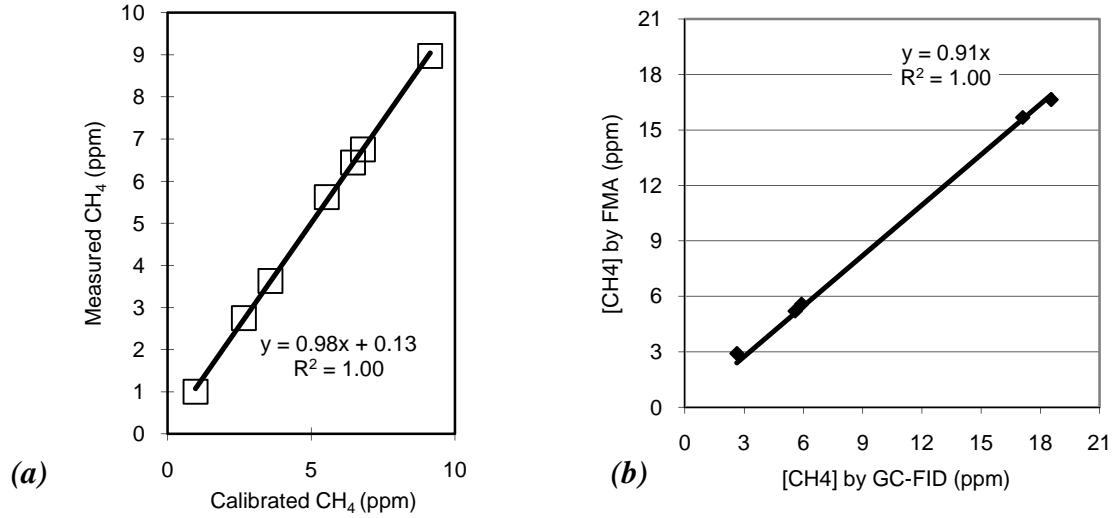


Figure 13 : Measurement of a reference standard gas with the FMA at the FFPRI (a). Cross-validation between the GC-FID and the FMA of the methane concentration measured at Yamashiro site on the 11th of July (b).

The values obtained with the GC-FID were 9% higher than those measured by the FMA but it may be due to the difficulties using a syringe for manual sampling from the soil chamber (*Brixie et al 1997*) or for collecting the high concentration calibration gas.

3.3. Temporal variations of the methane flux in the forest ecosystem

3.3.1. Seasonal variations

The methane flux was calculated from May to July, 21st. The quality controls led to omit about 20% of the data of the methane flux calculation.

In the chamber 1, located in dry conditions, the methane flux was almost constant and negative (*Figure 14*). In this study, it corresponded to a net flux uptake by the soil. The average of the methane flux from May to July was $-6.20 \cdot 10^{-3} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ ($\pm 1.10 \cdot 10^{-3} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$).

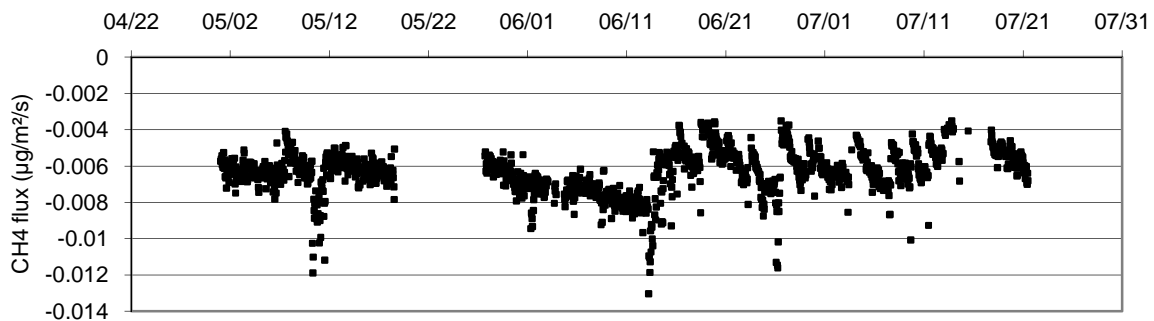


Figure 14 : The methane flux from the 1st of May to the 21st of July in the chamber 1 located in dry conditions after corrections by the quality controls.

The methane flux in the chamber 2 showed a particular pattern with a clear increase of the methane emission in June (*Figure 15*). In spring, the average of the methane flux emission was about $6.46 \cdot 10^{-4} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ ($\pm 2.45 \cdot 10^{-4} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$). In summer, the CH_4 flux increased up to $0.53 \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$, maximum value measured on the 15th of July at 2am.

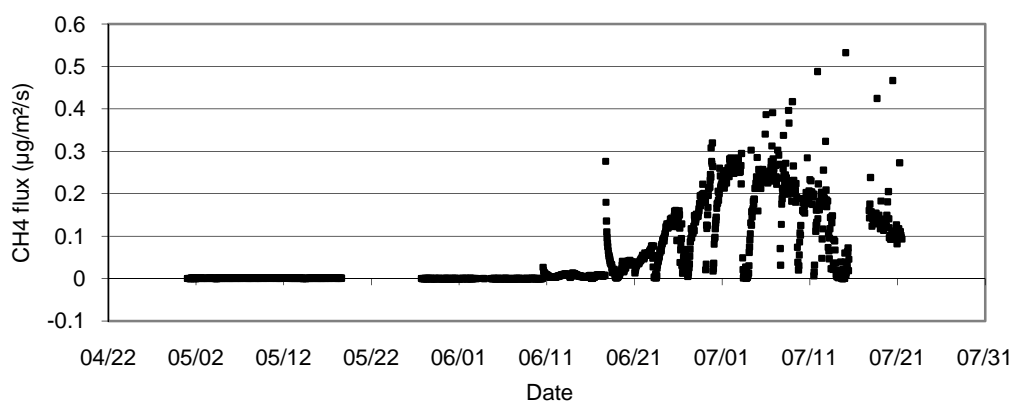
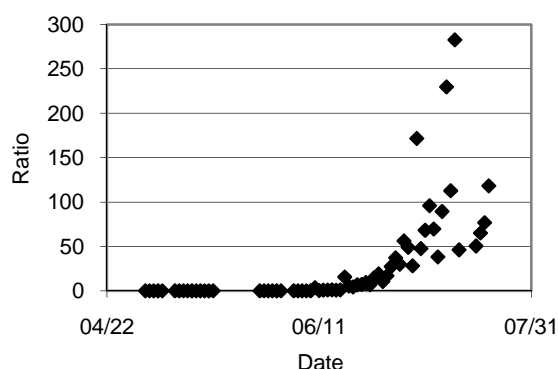


Figure 15 : The methane flux from the 1st of May to the 21st of July in the chamber 2 located in the river stream after corrections by the quality controls.



When the ratio $\frac{\text{emission}}{\text{uptake}}$ was calculated, the critical point between the stable and the increase of the net methane flux emission was found on the 24th of June (*Figure 16*). Moreover, between the spring and the summer patterns, the emission was more than 250 times higher than the uptake process.

Figure 16 : Ratio (uptake/emission) for the period of the study, calculated on the daily mean of the methane flux.

Last year, the methane concentration was measured at about the same place in the river stream using an automatic chamber (*Miyama et al 2010*). Air samples were collected once a week in a Tedlar bag and analyzed at the laboratory by GC-FID. The CH_4 measured was similar with the one estimated in this study (*Figure 17*).

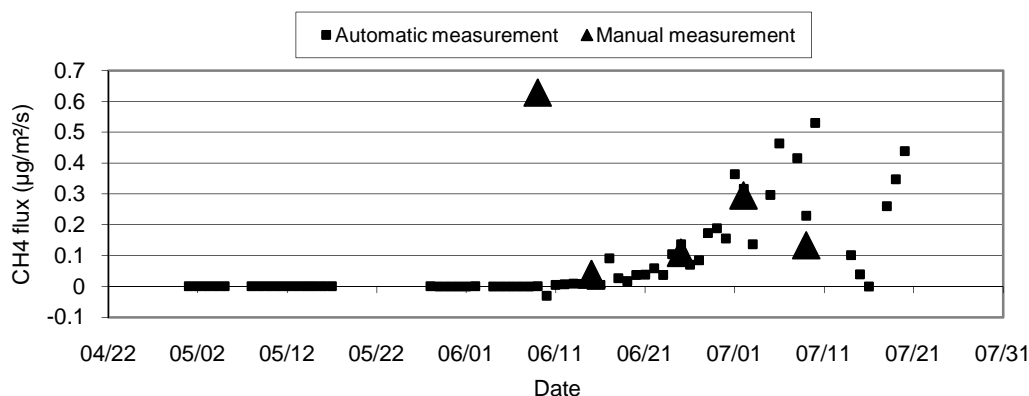


Figure 17 : Comparison of the manual and continuous measurements of the methane flux in the wet conditions in summer.

3.3.2. Variations with air temperature, precipitation and soil water content

The increase of the methane flux emission occurred later than the increase of the air temperature at YEF (*Figure 18*). In spring, the air temperature varied between 10 and 25°C whereas in summer the air temperature was always more than 20°C. The increase of the air temperature occurred at about the end of May, whereas the date of the change of the rate of the methane flux emission was about the mid of June. The correlation test between the air temperature and the methane flux showed a weak but significant correlation ($r^2 = 0.52$, P-value < 0.05).

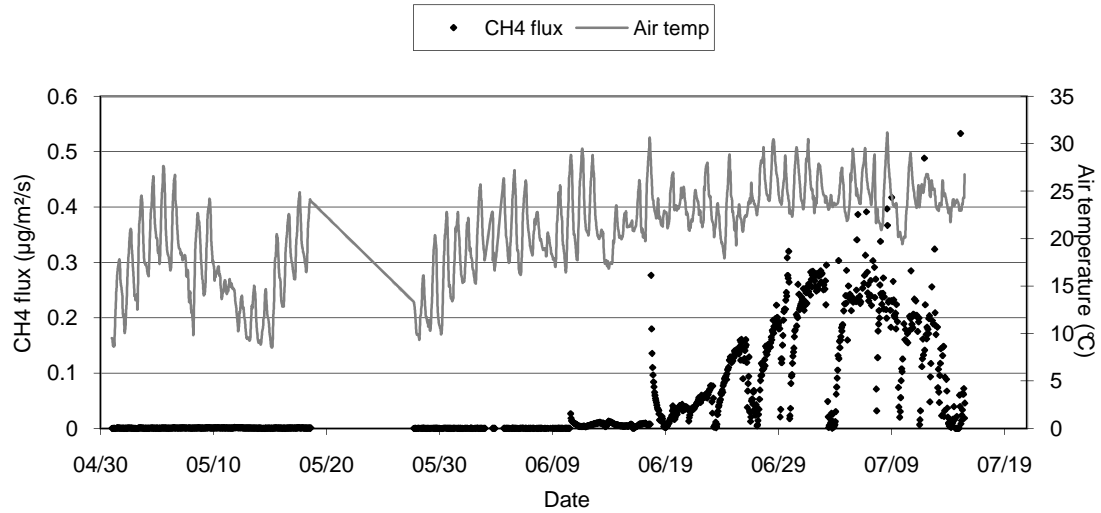


Figure 18 : CH₄ flux from the chamber 2 and air temperature at the ridge of the flux tower in the valley for the period of the study.

In Japan, a rainy season occurs from the end of June to the mid of July. At this period the CH₄ flux showed a pattern of increase / decrease at a daily scale. It seemed that the CH₄ flux decreased during a rainy event and then increased up to the previous value (*Figure 19*).

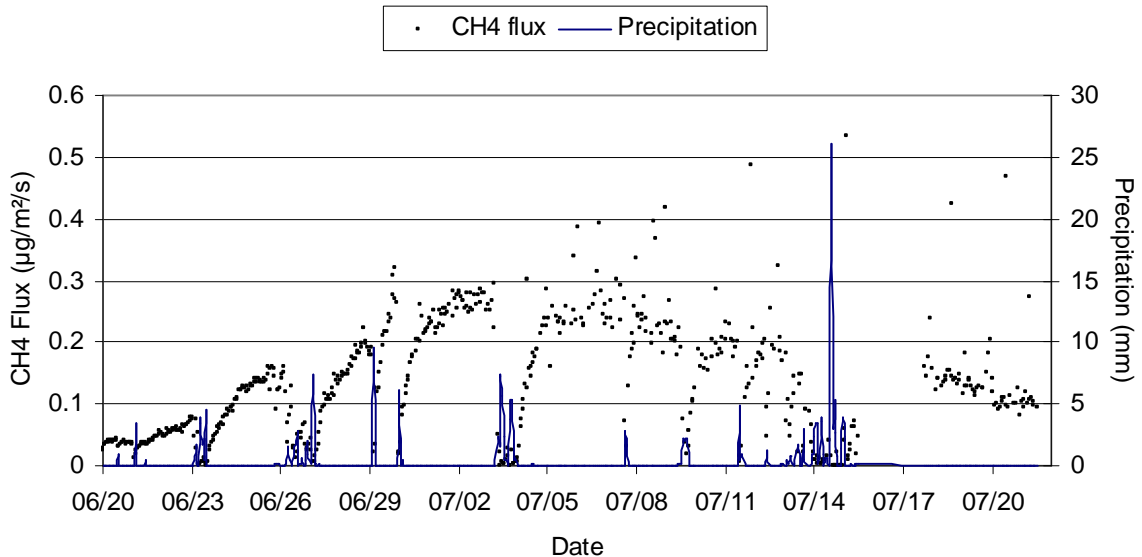


Figure 19 : CH₄ flux measured in the chamber 2 and precipitation from the mid of June to the 21st of July.

In details, the CH₄ flux emitted always decreased rapidly after a rainy period and then increased slowly (*Figure 20*). The CH₄ flux decreased of more than 90% in few hours (about 5 hours) and increased up to the previous in half a day.

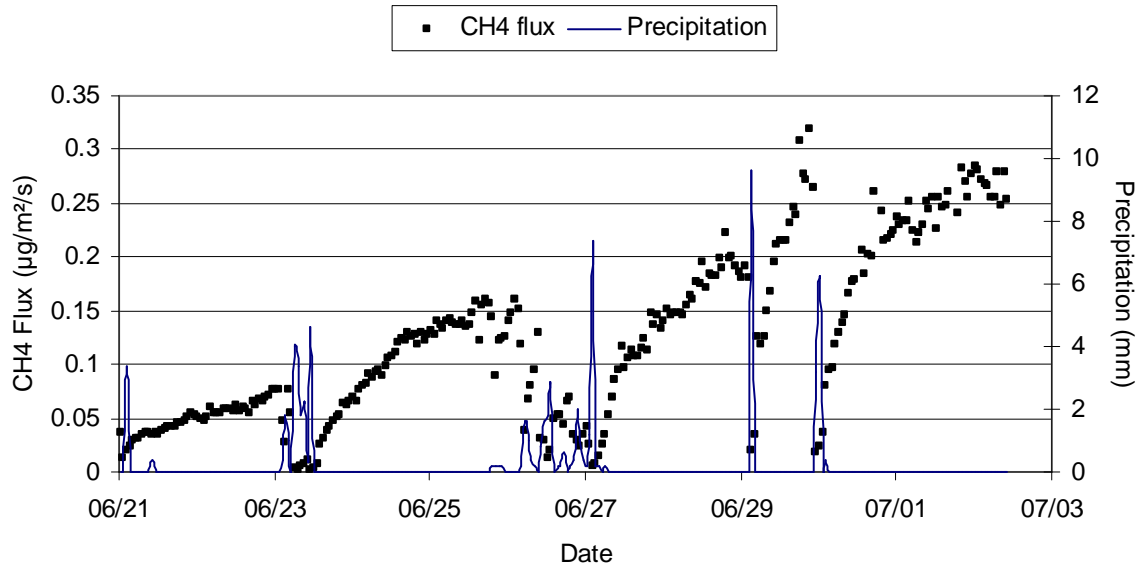


Figure 20 : CH₄ flux measured in the chamber 2 and precipitation from the 21st of June to the 2nd of July.

It was confirmed by the correlation test between the soil water content and the CH₄ flux calculated for the period of the study ($r = -0.80$, $P\text{-value} < 0.05$). In spite of the decrease of the CH₄ flux during a rainy event, the methane flux was always positive and the methane concentration always increased in the chamber 2.

3.3.3. Variations with the water temperature

The gas fluxes were correlated with the temperature of the water in the stream, especially for the CO₂ (Figure 21).

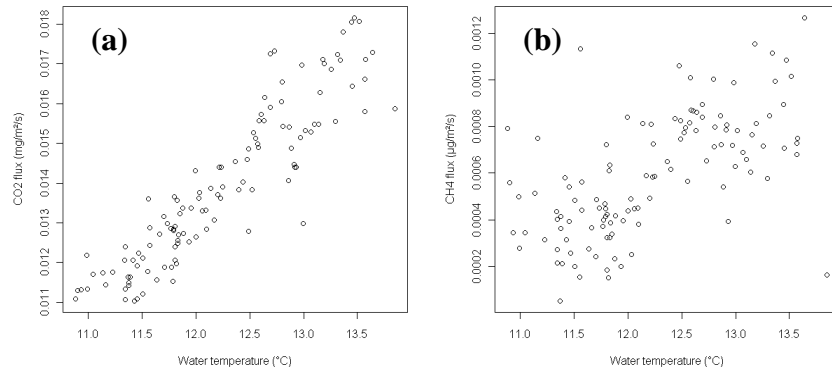


Figure 21 : The gas flux correlations in the chamber 2 from the 13th to the 18th of May. Correlation between the CO₂ flux and the water temperature (a, $r=0.91$, $P\text{-value} < 0.05$), between the CH₄ flux and the water temperature (b, $r=0.62$, $P\text{-value} < 0.05$)

After setting two heaters in the river stream under the chamber 2, the water temperature increased significantly, from 12°C to 30°C, the mean water temperature was 28.6°C in case of the 1st heater and 23.6°C in case of the 2nd heater. Each heater was set for one week. However, the methane flux did not increase. It may be due to the position of the heaters in the sediment or due to the rain event which occurred at those periods.

4. Discussion

4.1 The methane measurement using the FMA

In this study, we were able to measure the methane emitted or absorbed by the soil using the FMA developed by Los Gatos Research and based on an absorption-spectroscopy technique. The continuous measurement clearly shows a linear increase / decrease pattern of the methane concentration through the time. It allows the methane flux calculation using the slope of the linear regression and the time scale of the measurement. According to the first results obtained in YEF, the FMA is suitable to measure and to estimate the methane flux automatically and continuously. The cross-validation between the FMA and the GC-FID method validates the measurement by the FMA. The small differences of the absolute measured concentrations (9%) may be due to the transfer of the air using a syringe or at the standard deviation of the GC-FID. Moreover, the measurement of the carbon dioxide at the same time confirms the linear regression of the change of the concentrations and that there is no leak in the automatic sampling system.

The net absorption of the methane by the soil is constant through the seasons. In this study, the average of the methane absorption flux by the forest soil is $-6.20 \cdot 10^{-3} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ ($\pm 1.10 \cdot 10^{-3} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$). Morishita et al (2007) determined the mean CH_4 uptake rate at the forest floor in 26 sites throughout Japan over 2 years using a closed-chamber technique. They sampled and analyzed the methane concentration once a month by GC-FID. The annual mean rate of methane uptake reported is $-1.83 \cdot 10^{-2} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ (from $8.06 \cdot 10^{-4} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ to $4.86 \cdot 10^{-2} \mu\text{g} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$), which is similar to the results obtained by Miyama et al (2010) at the same site than in this study in YEF. The flux measured by the FMA is lower but it is difficult to compare such values because of the difference of the time scale. The annual mean integrates the large uptake as well as the weak absorptions by the soil in the same average.

The methane concentration measurement by an automatic multi chamber technique is fit for continuous measurement (Xunhua et al 1998). The little maintenance is required and the FMA can store the data almost indefinitely. The long distance transporting, the sample sucking with pumps and switch valves are not a source of error and the automatic system is reliable. However the measurement conducted in this study represents what happens above the chamber area in natural conditions. We can wonder if the stop of the water flow by the side of the chamber in the river stream would cause any disturbances in the soil dynamic. The water may be stagnant inside of the chamber and therefore create stable anoxic conditions which are favourable for the methanogenic bacteria (Yu et al 2008). Hence, the emission rate would be overestimated in spite of the accurate measurement by the FMA.

4.2 Temporal variability of the methane flux

As it is already reported in various papers, the forest ecosystem is a sink for the methane gas (Morishita et al 2007, Guckland et al 2009, Ishizuka et al 2009). But in this study, while a constant methane uptake rate was measured in dry conditions, a net methane emission was observed in wet conditions in the experimental temperate forest near Kyoto. Last year, a study was conducted to clarify the source of the methane emission at the same place than here in YEF. Air samples were collecting once a week using the closed-chamber method and analyzed at the laboratory by GC-FID. Strong peaks of methane emission were measured in summer in the chambers with a large amount of organic matter in the riparian zone. The continuous measurement with the FMA allows following the change, hour by hour, of the methane flux rate. In the chamber 2, a clear increase of the methane flux is observed in June, while it is the rainy season in Japan. The change can be clearly determined on the third week of June. From there, the rate of CH_4 production exceeds the CH_4 consumption and the

maximum value of the methane emission is 280 times higher than the uptake rate on the 13th of July. Then, the data show the beginning of a decrease of the methane flux intensity. One of the reasons of this decreasing pattern may be the short lifespan of the organic matter inside of the chamber. In Japan, the soil is very thin and because of the high humidity in summer, all the leaf litter accumulated in winter is rapidly decomposed in late summer. The high emission rate of the methane from the stream in the forest floor lasts about one month in summer, otherwise, the net methane emission flux is weak but it still remains. In this case the forest floor acts as a source of methane. It may be due to the stability of the favourable conditions required by the methanogenic bacteria in the sediment in the river stream. This finding shows the potential of the forest ecosystem in emitting the methane gas into the atmosphere and it could make us revalue the global methane circulation at a global scale. However, further studies are required to explain those favourable conditions: what are they? How do they control the methane emission?

In spite of a clear seasonal pattern, the methane flux does not show any daily pattern. However, the FMA measures the methane concentration every second and it will be interested to analyze the data for one year by integrating the methane flux measured hourly, daily and seasonally.

4.3 Environmental factors influencing the methane flux

The methane flux results from the balance between the methane emitted into the atmosphere and the methane absorbed into the soil. The net methane flux can be positive or negative according to the process which is dominant (consumption or emission by the soil). The reasons of the fluctuations of the methane flux into the soil are still unclear. It seems that the methane cycle is correlated with some environmental factors, like the water content, but there is no evidence yet.

In Yamashiro site, the methane emission increases at the end of spring and during the rainy season which correspond to a change in the environmental conditions. The flow rate becomes stronger because of the heavy rain, the soil water content increase as well as the air temperature. The methanogenic bacteria may be stimulated by the air temperature (*Le Mer et al 2001*) but it is not the main reason. The methane flux in the forest soil is a complex dynamic and it is not realistic to analyze the methane flux according to only one environmental factor, especially for the CH₄ (*Arnold et al 2005*). More data about the oxygenation in the river stream or the water level are required. Moreover, it was expected that the water temperature had an influence on the methane emission but no correlation is found between the methane emission and the increasing temperature of the water inside the chamber 2. The water temperature superior to 25°C might be too high for the methanogenic bacteria. On the contrary, the carbon dioxide emission increases with the water temperature, it is due to the less solubility of this gas with the increasing temperature (*Luo et al 2006*).

At a smaller scale, an increase of the soil water content marks a heavy rain and a decrease of the methane emission in the chamber 2. A rainy event disturbs the stability of the conditions in the forest floor and so inside of the chamber. A high flow rate may increase the amount of oxygen and stop the methanogenic activity because of the sensitivity of methanogens to the oxygen (*Megonigal et al 2008*). On the other hand, methanogens and methanotrophs are present simultaneously (*Le Mer et al 2001*) and the aerobic conditions may create good conditions for methanotrophic bacteria. Indeed, methanotrophs are inhibited by diffusional limitation of CH₄ through the water (*Bradford et al 2001*). The decrease of the methane emission flux is very fast which suggests a high reactivity of the bacteria to the change of the environmental factors.

5. Conclusion

The first results obtained from April to July 2010 validate the utilization of the Fast Methane Analyzer in the gas flux estimation in the forest soil studies. The low concentration of the methane in the atmosphere is detected by the analyzer and the slope of the decreasing/increasing pattern can be used to calculate the flux. Once the system is set in the field site, the FMA is easy to use and does not need any particular maintenance. The methane concentration is calculated according to the cell adjustment and does not need to be calibrated. Using the FMA in a forest site is suitable, even at very low or very high temperatures as it occurs in Kyoto. The cross validation using the gas chromatograph equipped with a flame ionization detector confirmed the CH₄ concentration measured by the FMA. The methane concentration is the same order, as well as the methane flux from the river which was estimated for many years using the GC method.

However, the FMA can not totally replace the GC-FID measurement because only a few numbers of automatic chambers can be connected. It is not enough to measure the spatial variations of the methane flux in the forest floor. For instance, in Yamashiro Experimental Forest, the manual method, using Tedlar bags, is conducted on eight soil chambers from three different places along the river. In brief, both manual and automatic samplings have disadvantages and advantages and may be used simultaneously. The automatic chambers give information about the temporal variation whereas the manual method provides knowledge about the spatial variation of the methane flux in the forest ecosystem.

The characteristics and factors of the spatial and temporal variation in methane concentration in temperate forest soils are still unclear (*Miyama et al 2010*). Many factors influence the CH₄ dynamic. This study focuses on the setting of the fast methane analyzer but, after the complete understanding of this new tool, the study of the environmental parameters adding to the continuous measurement will provide some information and more knowledge about the methane flux in the forest ecosystem.

The measurement of the methane flux in the forest ecosystem is a key of the methane cycle understanding. The continuous analyze is a tool to follow the changes of the gas concentration in the atmosphere, especially about the greenhouse gases. And, in the context of the climate change, we can wonder if the change of the net methane emission rate in June will come sooner year after year. As well as the climate change modifies the process of decomposition and the amount of leaf litter, we can wonder if the methane emission rate will be affected.

Bibliography

- Akiyama H., Hayakawa A., Sudo S., Yonemura S., Tanonaka T. & Yagi K., 2009. Automated sampling system for long-term monitoring of nitrous oxide and methane fluxes from soils. *Soil Science and Plant Nutrition*. 55: 435–440
- Arnold K., Nilsson M., Hanell B., Weslien P. & Klemmedtsson L., 2005. Fluxes of CO₂, CH₄ and N₂O from drained organic soils in deciduous forests. *Soil Biology & Biochemistry*. 37: 1059–1071
- Baer D.S., Paul J.B., Gupta M. & O’Keefe A., 2002. Sensitive absorption measurements in the near-infrared region using off-axis integrated-cavity-output-spectroscopy. *Applied physics B*. 75: 261–265
- Borken W., Davidson E.A., Savage K., Sundquist E.T., Steudler P., 2006. Effect of summer throughfall exclusion, summer drought, and winter snow cover on methane fluxes in a temperate forest soil. *Soil Biology & Biochemistry*. 38: 1388–1395
- Bradford M.A., Ineson P., Wookey P.A. & Lappin-Scott H.M., 2001. Role of CH₄ oxidation, production and transport in forest soil CH₄ flux. *Soil Biology & Biochemistry*. 33: 1625–1631
- Brixie J., Neilson L., Kim B. & Severin A., 1997. Bottle-in-bag technique for gas sampling with analysis by autoheadspace GC. *Environmental science & Technology*. 31: 1224–1228
- Dlugokencky E.J., Masarie K.A., Tans P.P., Conway T.J. & Xiong X., 1997. Is the amplitude of the methane seasonal cycle changing ? *Atmospheric Environment*. 31: 21–26
- Evans J.R., 2007. Resolving methane fluxes. *New Phytologist*. 175: 1–4
- GIEC, 2007. *Bilan 2007 des changements climatiques. Contribution des Groupes de travail I, II et III au quatrième Rapport d’évaluation du Groupe d’experts intergouvernemental sur l’évolution du climat*. Publié sous la direction de Pachauri R.K. et Reisinger A. GIEC, Genève, Suisse, 103 pages
- Guckland A., Flessa H. & Prenzel J., 2009. Controls of temporal and spatial variability of methane uptake in soils of a temperate deciduous forest with different abundance of European beech (*Fagus sylvatica* L.). *Soil Biology & Biochemistry*. 41: 1659–1667
- Hendriks D.M.D., Dolman A.J., Van der Molen M.K. and Van Huissteden J., 2008. A compact and stable eddy covariance set-up for methane measurements using off-axis integrated cavity output spectroscopy. *Atmospheric Chemistry and Physics*. 8: 431–443
- Ishizuka S., Sakata T., Sawata S., Ikeda S., Sakai H., Takenaka C., Tamai N., Onodera S., Shimizu T., Kan-na K., Tanaka N. & Takahashi M., 2009. Methane uptake rates in Japanese forest soils depend on the oxidation ability of topsoil, with a new estimate for global methane uptake in temperate forest. *Biogeochemistry*. 92: 281–295
- Ishizuka S., Sakata T. & Ishizuka K., 2000. Methane oxidation in Japanese forest soils. *Soil Biology & Biochemistry*. 32: 769–777
- Jomura M., Kominami Y., Tamai Y., Miyama T., Goto Y., Dannoura M. & Kanazawa Y., 2007. The carbon budget of coarse woody debris in a temperate broad-leaved secondary forest in Japan. *Tellus*. 59: 211–222
- Le Mer J. & Roger P., 2001. Production, oxidation, emission and consumption of methane by soils: A review. *Eur. J. Soil Biol.* 37: 25–50
- Los Gatos Research, 2009. <http://www.lgrinc.com/>

- Luo Y. & Zhou X., 2006. Soil respiration and the environment. USA: Elsevier, 316p
- Miyama T., Hashimoto T., Kominami Y., Nakagawa K., Okumura M. and Tohno S., 2010. Temporal and spatial variations in CH₄ concentrations in a Japanese warm-temperate mixed forest. *Agricultural and Forest Meteorology*. 66: 1-9
- Megonigal P.J. & Guenther A.B., 2008. Methane emissions from upland forest soils and vegetation. *Tree Physiology*. 28: 491-498
- Menyailo O.V. & Hungate B.A., 2003. Interactive effects of tree species and soil moisture on methane consumption. *Soil Biology & Biochemistry*. 35: 625–628
- Morishita T., Sakata T., Takahashi M., Ishizuka S., Mizoguchi T., Inagaki Y., Terazawa K., Sawata S., Igarashi M., Yasuda H., Koyama Y., Suzuki H., Toyota N., Muro M., Kinjo M., Yamamoto H., Ashiya D., Kanazawa Y., Hashimoto T. and Umata T., 2007. Methane uptake and nitrous oxide emission in Japanese forest soils and their relations to soil and vegetation types. *Soil Science and Plant Nutrition*. 53: 678-691
- Perma pure LLC, <http://www.permapure.com/>
- Picarro, 2010. <http://www.picarro.com/>
- Schnurr-Pütz S., Baath E., Guggenberger G., Drake H.L. & Küsel K., 2006. Compaction of forest soil by logging machinery favours occurrence of prokaryotes. *FEMS Microbiol Ecol*. 58: 503–516
- Tamai N., Takenaka C., Ishizuka S. & Tezuka T., 2003. Methane flux and regulatory variables in soils of three equal-aged Japanese cypress (*Chamaecyparis obtusa*) forests in central Japan. *Soil Biology & Biochemistry*. 35: 633–641
- Tan Z. & Long X., 2010. Off-axis integrated cavity output spectroscopy and its application. *Optics communications*. 238: 1406-1409
- Xunhua Z., Mingxing W., Yuesi W., Renxing S., Jing L., Heyer J., Kogge M., Laotu L. and Jisheng J., 1998. Comparison of manual and automatic methods for measurement of methane emission from rice paddy fields. *Advances in atmospheric sciences*. 14: 569:579
- Yao Z., Zheng X., Xie B., Liu C., Mei B., Dong H., Butterbach-Bahl K. & Zhu J., 2009. Comparison of manual and automated chambers for field measurements of N₂O, CH₄, CO₂ fluxes from cultivated land. *Atmospheric Environment*. 43: 1888–1896
- Yu K., Faulkner S.P. & Baldwin M.J., 2008. Effect of hydrological conditions on nitrous oxide, methane, and carbon dioxide dynamics in a bottomland hardwood forest and its implication for soil carbon sequestration. *Global Change Biology*. 14: 798–812