

Methane Emission and Consumption in Russian *Gray Forest* Agricultural Soil: Monthly, Daily and Diurnal Variations and Effect of Mineral and Organic Fertilization

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Abstract: *Gray forest* soils are one of the typical agricultural soils in European part of Russia, but their role as a sink for atmospheric methane is poorly documented. We have investigated variability of CH₄ uptake process of agricultural *gray forest* soils, Moscow region, Russia, at different time scales (monthly, daily, hourly). Additionally some belowground factors impacting soil CH₄ consumption were tested for their short- and long-term effect. It was found that the *gray forest* soil was the sink for atmospheric methane and average annual net rate of methane oxidation for 3 years was calculated as 20 mcg C-CH₄ m⁻² h⁻¹. The large fluctuations were evaluated in response to the daily cycle and were found to be strongly influenced by air temperature. Nitrogen and manure fertilization resulted in short-term (3-4 weeks) shift from CH₄ uptake to CH₄ emission, which was determined by transformation of labile fractions of soil organic matter.

Key words: Soils • Methane uptake • Time scales • Nitrogen and manure fertilization

INTRODUCTION

Increases in the concentrations of methane (CH₄), important atmospheric greenhouse gas, due to human activities are associated with global climate change. A significant sink exists for CH₄ (about 6%) in aerobic soils, including forestry and agricultural soils [1]. Many studies have investigated CH₄ uptake in soils of arable cropping ecosystems and have found them to be the sink for atmospheric methane [2-4]. At the same time the rates of CH₄ consumption in arable soils is generally lower than from native ecosystems under similar environmental conditions [2, 3] and usually are only about 10- 30% of those under forest. Until now very limited data are available for methane oxidation and consumption in Russian agricultural soils. The only regular in situ research was done in Moscow region [5] and extrapolated for all *gray*

forest soils in this region the CH₄ fluxes gave negative annual fluxes of 2.3 kg C-CH₄ ha⁻¹ year⁻¹ for native forest and 0.8 kg for wheat agrocenosis. *Gray forest* soils cover about 60 million of ha or around 2.8 % of Russia's landscape and about 40% of them are arable cropping. So, monitoring and process understanding of CH₄ consumption in these soils is required to estimate global greenhouse gases balance and contribution to global warming.

Since the Russian climate and seasonality is extremely variable, CH₄ consumption is spatially and temporally variable and temporal variability exists in many scales with different controlling factors. Recently we have determined significant seasonal variations in soil CH₄ fluxes from Russian *gray forest* soils under different ecosystems (native forest, arable cropping) and have evaluated that these differences governed by environmental regulators [5].

Numerous investigations were conducted in order to evaluate factors controlling CH_4 oxidation and consumption. It was shown that diurnal variations may be more than threefold and cause systematic errors in CH_4 flux estimations [6]. The effect of temperature is inconsistent and usually small. In forest soils soil temperature was found to be an important controller of CH_4 consumption at temperatures between -5 and 10°C , but had very little affect at 10 and 20°C [7]. The Q_{10} values for CH_4 oxidation range from 1.1 to 4.8 [8]. Methane oxidation negatively correlated with soil water content, probably due to limited CH_4 diffusion [9]. However, net CH_4 oxidation is possible in wet soil due to localized aerobic microsites or anaerobic CH_4 oxidation [10].

Ammonium fertilization is usually reported to competitively inhibit CH_4 oxidation [11; 12]. Some studies, however, reported no effect [13] or positive promotion [14]. These conflicting effects may be due to diversity in methanotrophic communities. It was revealed that application of N fertilizers generally inhibits methane oxidation by type II methanotrophs, but enhances by type I methanotrophs. As such, the co-existence of different methane-oxidizing communities may reduce the inhibitory effect of NH_4^+ on methane oxidation [15]. NO_3^- inhibits CH_4 oxidation in arable soils [15], but persists in soil only for a few days. The effect of NO_3^- is unclear and varies from either no effect [13] to negative effect [16].

Organic amendments vary in their effects on CH_4 consumption according to their C: N ratios. For example, adding fresh sugar beet leaves reduced CH_4 consumption to 20%, whereas adding wheat straw had no effect [17]. Long-term application of organic fertilizers supports diversity of methanotrophs, as was estimated by group-specific DGGE of 16S rRNA genes [18].

For this study we have opportunity during 4 years to investigate soil CH_4 fluxes in experimental micro-plots of *gray forest* soil with different agricultural management in

Experimental Pavilion of IPCBPSS RAS. We hypothesized that methane flux is follows not only important inter-annual variations, but demonstrated daily and diurnal changes. The objectives were to reveal and compare variations of CH_4 consumption in different temporal scales and fertilization and characterize the relative importance of environmental factors (e.g. temperature, soil moisture, fertilization) on magnitude of methane flux.

MATERIALS AND METHODS

Site Description: The study was conducted 2001-2003 and 2005 at Experimental Pavilion of Institute of Physicochemical and Biological Problems in Soil Science (IPCBPP), Russian Academy of Sciences which is located in European part of Russia, 150 km to S from Moscow ($54^\circ50'\text{N}$, $37^\circ35'\text{E}$). The average annual precipitation is 582 mm and the mean annual temperature is $+3.9^\circ\text{C}$, but with great variability within seasons as well as between years. The experimental site consisted of individual 0.25 m^2 A-micro-plots and 4 m^2 B-micro-plots, each lined by 5-cm thick concrete walls. The soil was moderately eroded gray forest soils (humic luvisols) with pH ranged from 5.8 to 6.5, organic C 0.96%, total N 0.1%, N_{min} 8.2 mg kg^{-1} .

Four separate experiments were conducted and the following treatments are listed at Table 1. Samples for analytical determinations were taken in the center of each micro-plot in triplicate at monthly intervals.

Analysis of Soil CH_4 Fluxes and Monitoring

Environmental Factors: Carbon flux from the soil surface, as CH_4 evolved, was monitored using the static chamber method with gas chromatography (GC) analysis [5] with one measuring point in each micro-plot (total $n = 9$ or 12). CH_4 fluxes were measured 1, 6, 18, 33 and 47 days after N-fertilization (Experiment I), 33, 62, 78, 93, 105, 133 and 148 days after green manure application.

Table 1: Experimental design of the four micro-plot experiments

	I			II			III			IV		
Year	2001			2002			2003			2005		
Experiment	I-1	I-2	I-3	II-1	II-2	II-3	III-1	III-2	III-3	IV-1	IV-2	IV-3
Culture Fertilizer	Unplanted	Unplanted	Oat	Unplanted	Unplanted	Oat	Unplanted	Unplanted	Corn	lack radish	lack radish	Semi-natural
treatment	fallow	fallow $\text{Ca}(\text{NO}_3)_2^*$	$\text{Ca}(\text{NO}_3)_2$	fallow	fallow	Green	fallow	fallow	Green	Traditional	Organic	bluegrass
					Green	manure		Green	manure	fertilization	fertilization	grassland
						manure**		manure		system***	system****	

* 7.5 g m^{-2} in the form of water solution was injected into soil at May 2001.

** Ground top oat biomass from Experiment I-3; 0.1 kg m^{-2} was incorporated into upper 0-20 cm layer at May 2002 (Experiment II) and October 2002 (Experiment III); $\text{N}_{\text{tot}}=1.59\%$; C:N=26

*** 10 g m^{-2} N, P_2O_5 , K_2O and 0.25 g m^{-2} herbicide 2,4,D

**** 2.5 kg m^{-2} of fresh cattle manure; $\text{C}_{\text{org}}=36.5\%$; $\text{N}_{\text{tot}}=1.63\%$

Weather and soil conditions were estimated simultaneously with flux measurements. Soil moisture content was measured with Theta probe (HM Digital), pH with Piccolo plus (Hanna) pH-meter. Two weather stations (Oregon Scientific) were installed on the soil surface to monitor air temperature, pressure and humidity and soil temperature sensors (Fisher Scientific) were installed at 10 cm depth.

Analytical Procedures and Statistical Analysis: For a statistical analysis of CH_4 fluxes, environmental and soil parameters, simple and multivariate correlations and standard errors were calculated with statistical procedures.

RESULTS AND DISCUSSION

Effect of N-fertilization, Oat Biomass Incorporation and Cropping on Monthly Fluctuations of CH_4 Fluxes: Soil fluxes were determined at irregular intervals during three growing periods (Fig. 1). Methane fluxes in fallow micro-plots were negative in summer time and positive in autumn (Fig. 1, Exp.II-1) and spring (Exp. III-1). The application of nitrogen fertilizer and oat cropping resulted in short-term inhibition of methane uptake, which was demonstrated as positive fluxes (Fig. 1, Exp. I-2 and I-3). After 3 months we did not observe any significant difference in CH_4 flux between variants ($P>0.05$).

Application of plant residues at the beginning of growing season have resulted in appreciable increase of CH_4 uptake in fallow plots, which was up to $0.298 \text{ mg C-CH}_4 \text{ m}^{-2} \text{ h}^{-1}$ in variant II-2 comparably with $0.069 \text{ mg C-CH}_4 \text{ m}^{-2} \text{ h}^{-1}$ in variant II-1 at the same time. This effect was preserved when the plant residues were incorporated into soil in autumn (Exp. III) and at the next year the extent of increase in CH_4 uptake was practically the same – 2-5 times. When the oat was cropping on N-fertilized plots (Exp. II-3) the differences in flux were not statistically different ($P>0.05$) from fallow. The same results were obtained for maize cropping experiment (Exp. III-3) with fallow and fallow with green manure.

Diurnal Fluctuations of CH_4 Flux: We measured changes of CH_4 flux within a day in Exp.III in spring, summer and autumn (Table 2) and have found large diurnal variability. For example, in fallow micro-plots the measured values ranged from -0.017 to $0.026 \text{ mg C-CH}_4 \text{ m}^{-2} \text{ h}^{-1}$ in April, from -0.032 to 0.009 mg in July and from -0.024 to 0.006 mg in September. Diurnal rhythm was similar in micro-plots with different management and they showed absorption peak at 15-00. At night time CH_4 uptake had decreased greatly in all micro-plots and they showed emission. The average soil fluxes from the measurements between 9:00 and 10:00 can be regarded as the representative of daily averages.

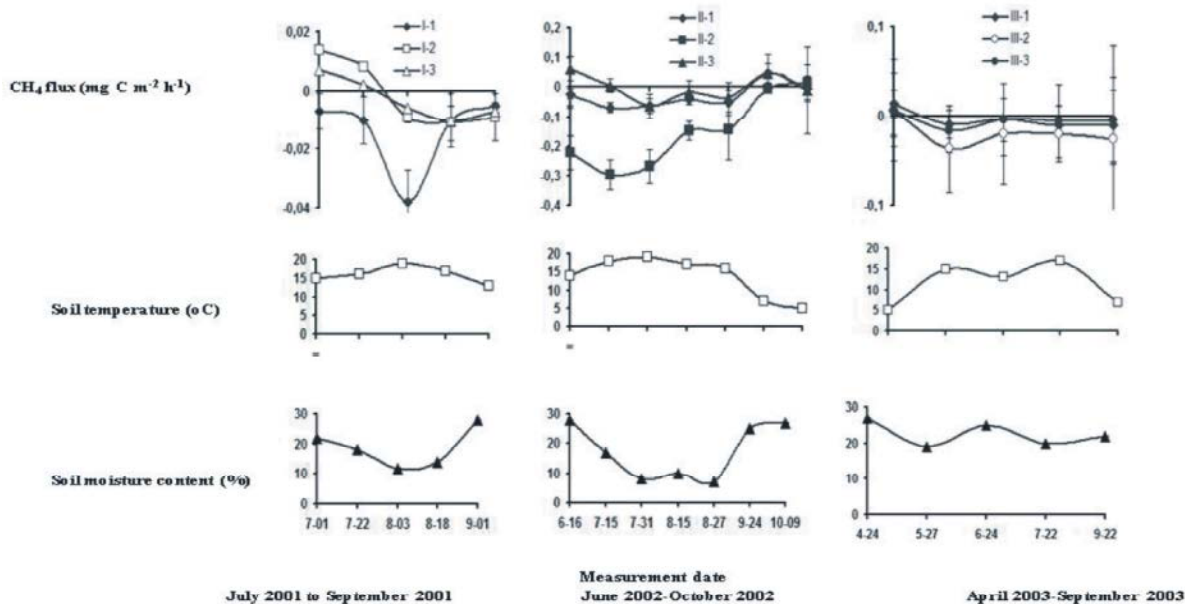


Fig. 1: Monthly trends in measured CH_4 fluxes, soil temperature and soil moisture for growing seasons 2001 (Experiment I), 2002 (Experiment II) and 2003 (Experiment III) in micro-plots of experimental sites with different treatments. Vertical bars are standard errors of the mean ($n=3$) for each sampling day.

Table 2: Diurnal oscillations of methane flux in Exp.III, mg C-CH₄m⁻² h⁻¹

Date of sampling	Variant	Time of Day			
		9 a.m.	3 p.m.	9 p.m.	3 a.m.
April 24	7	0.014±0.004	- 0.017±0.081	- 0.014±0.004	0.026±0.012
	8	0.006±0.000	- 0.055±0.093	- 0.040±0.073	- 0.003±0.004
	Average	0.010±0.005	- 0.036±0.075	- 0.027±0.045	0.011±0.018
July 22	7	- 0.011±0.000	- 0.032±0.004	- 0.011±0.008	0.009±0.012
	8	- 0.020±0.004	- 0.043±0.012	- 0.020±0.004	- 0.011±0.000
	9	- 0.006±0.000	- 0.010±0.018	0.014±0.012	0.023±0.016
	Average	- 0.012±0.007	- 0.028±0.018	-0.006±0.017	0.007±0.018
September 22	7	- 0.011±0.000	- 0.024±0.002	0.014±0.004	0.006±0.024
	8	- 0.026±0.004	- 0.031±0.022	- 0.020±0.045	0.014±0.004
	9	- 0.005±0.010	- 0.014±0.020	- 0.009±0.004	0.020±0.004
	Average	- 0.014±0.011	- 0.023±0.015	- 0.014±0.021	0.013±0.013
Total average	- 0.007±0.013	- 0.028±0.036	- 0.014±0.027	0.010±0.015	

The “±” indicate the standard error of mean

Table 3: Correlation analysis of methane fluxes and environmental factors

Experiment #	Number of estimations	Correlation coefficient		
		Air temperature	Soil temperature	Soil moisture
I	15	NS	NS	NS
II	21	-0.515*	-0.518*	0.436*
III	14	NS	NS	0.634*
III*	32	-0.504**	NS	NS
IV	75	NS	NS	0.288**
Total	125	-0.238**	-0.271**	0.290***

Note: All estimations were done at 9-10 a.m. period with the exception of III*, when estimations were done at different time.

NS- not significant; significant at P-level < 0.05*, <0.01**, <0.001***

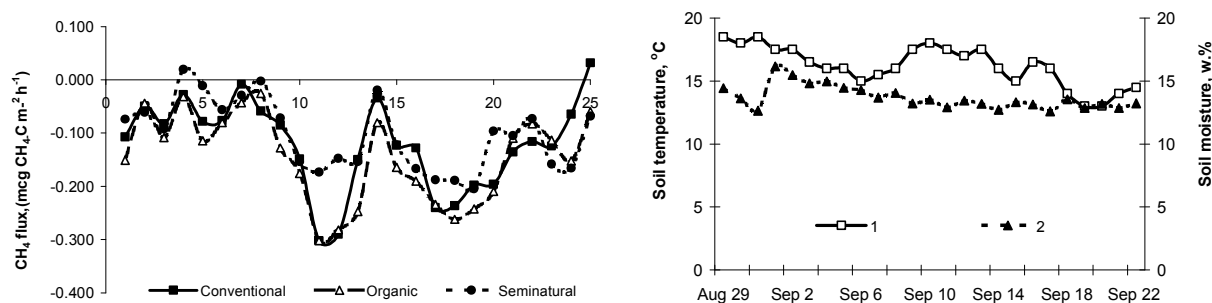


Fig. 2: Daily variations in measured mean soil CH₄ fluxes (n=3) from micro-plots of Experiment IV (a) and soil temperature and moisture (b).

Effect of Traditional and Organic Fertilization on CH₄ Fluxes: In this experiment soil flux was determined every day at 9-00 from 29 August to 22 September 2005 (25 days). It was found, that both fertilization systems and cropping of black radish in summer (July) did not affect significantly the average CH₄ flux value ($P > 0.05$). The every day time series of CH₄ fluxes have revealed important wake-like variations (Fig. 2). During the observation period daily average fluxes ranged from two to twelve fold at 2-4 days apart.

Relations Between Soil Temperature, Soil Moisture and CH₄ Fluxes: Correlation analysis revealed a good relationship between CH₄ flux and soil temperature and moisture (Table 3).

Both mean soil temperature and mean air temperature were significantly ($P = 0.05$) correlated (R^2 ranging from 0.84 to 0.92) with CH₄ flux regardless of whether mean hourly or mean daily rates were used. Mean air temperatures were also highly correlated with soil temperature as seen from the correlation coefficient (R^2) of

0.90 ($P < 0.001$). Soil moisture, however, varied relatively little, but strongly affected methane flux rates at monthly and daily scales. Conversely, under the conditions of this study, it was predominantly temperatures that affected soil biological activity and hence, soil methane uptake at hourly rates ($R^2=0.82$, $P < 0.001$). Regression analyses revealed that a number of linear and nonlinear functions predicted the effect of soil temperature and moisture reasonably well for monthly, diurnal and hourly flux rates (equations not shown). Best-fit relationships were provided by second-order polynomial curves.

CONCLUSIONS

Gray forest soils, one of the typical agricultural soils in European part of Russia, are important participants in the global carbon budget, but their role as a sink for atmospheric methane is poorly documented. We have investigated variability of CH_4 uptake process and environmental and edaphic characteristics of agricultural *gray forest* soils, Moscow region, Russia, at different time scales (monthly, daily, hourly). Soil CH_4 uptake was measured using a closed chamber method to quantify soil flux and to determine the contribution of soil temperature, soil moisture and dissolved organic carbon and nitrogen content. The field data were analyzed in order to elucidate the mechanisms governing short-term and long-term trends and try to illuminate what we miss if we monitor or model this process coarsely in time. Additionally some belowground factors impacting soil CH_4 consumption, namely mineral nitrogen compounds, wheat straw and manure treatment, were tested for their short- and long-term effect.

The following general conclusions can be drawn from this study:

- In total the *gray forest* soil was the sink for atmospheric methane and average net rate of CH_4 oxidation for 3 years was calculated as $20 \text{ mcg C-CH}_4 \text{ m}^{-2} \text{ h}^{-1}$.
- The large fluctuations were evaluated in response to the daily cycle and CH_4 uptake was registered at day-time, but emission at night-time. The average soil fluxes measurements between 9:00 and 10:00 can be regarded as the representative of daily averages. These diurnal variations were found to be strongly influenced by air temperature and Q_{10} for methane uptake by surface soil was found to be 2.5 ± 0.5 .
- If intraday fluctuations in CH_4 fluxes are ignored and mean values are used, measurements may result gross overestimates, underestimates, or even the wrong sign of process.
- Nitrogen and manure fertilization resulted in short-term (3-4 weeks) shift from CH_4 uptake to CH_4 emission, which was determined by transformation of labile fractions of soil organic matter.

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