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Measurements of methane fluxes in the surface layer of the atmosphere over Western Siberia

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ABSTRACT

The paper presents the results of the estimations of vertical methane fluxes obtained by using the gradient method. The gradient measurements of methane concentration and meteorological parameters have been obtained from the Observatory Fonovaya of V.E. Zuev Atmospheric optics institute SB RAS from July 2016 to December 2018

Keywords: vertical methane fluxes, surface layer, measuring mast

1. INTRODUCTION

Methane (CH4) in the atmosphere is the second most important greenhouse gas after carbon dioxide. The balance of sources and sinks determines CH4 concentration. Emission sources comprise anthropogenic activity fossil fuel combustion, rice agriculture, livestock, landfill, and waste treatment, and some biomass burning and natural sources such as wetlands, termites and the ocean. Two processes determine CH4 sinks: absorption by soils (methanotrophs) and oxidation in the atmosphere during reactions with the hydroxyl radical (OH). Estimations of methane sinks in the atmosphere (especially on reactions with OH) are complicated. The vertical fluxes and the rate of dry deposition are used to estimate sources and sinks. The work aims to study the vertical flux of methane for the background area of Western Siberia.

2. MEASUREMENT DATA AND CALCULATION METHODS

Based on the measurements of methane concentration and meteorological parameters has been derived from the altitude mast installed in the Fonovaya Observatory of IAO SB RAS (August 2015-December 2018), the calculation of the vertical methane fluxes was performed. The Fonovaya Observatory is located in the background region of the Tomsk Region (56°25' N, 84°04' E, 80 m. above the sea level, (the current condition available at http://lop.iao.ru). The list of measuring equipment is shown in Table 1. Methane concentrations were measured at 10 and 44 m (lower and upper levels respectively), meteorological parameters were measured at 10 and 40 m. Meteorological parameters were recorded every hour. The gas composition was measured in the following way. From 0 to 30 minutes, the concentration was measured from the upper level, from 30 to 60 minutes from the lower level. To agreement the measurement time, linear interpolation between adjacent measurement times was used. The concentration values obtained for the upper level were interpolated.

The vertical fluxes of carbon dioxide and methane were calculated by the gradient technique based on the MoninObukhov similarity theory, which is described in detail in [1]. According to this technique, the gas flux is calculated by Eq. (1) as

$$F_s(z_m) = -K\frac{\partial S}{\partial z} \tag{1}$$

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Table 1. Equipment of Fonovaya Observatory.

Device/sensor	Parameter	Range	Error	Time constant
Vaisala HMP155	t °C	-40...+60	± 0.1 C	1 s
Vaisala HMP155	f,%	0...100	±2%	1 s
Young model 85004	dd, deg	0...360	± 10°	1 s
Young model 85004	V, m/s	1,2...40	±(0.5+0.05V)	1 s
Young model 61302V	P, hPa	150...1150	± 1.5	0.1 s
Picarro G2301-m	CO_2, ppm	0...1000	< 0,2 ppm	1 s
	CH_4, ppm	0...20	< 0.0015 ppm	1 s
	H_2O, ppm	0...70000	< 150 ppm	1 s

where $F_s(z_m)$ is the vertical flux of a substance, K is the turbulent diffusion coefficient, $\frac{\partial S}{\partial z}$ is the gradient of the substance concentration, $z_m = \sqrt{z_1 z_2}$ is a reference height, for which the flux is calculated, $z_2 = 40$ and $z_1 = 10$ are the heights of the upper and lower measurement levels, respectively.

The following rule is used in the calculations: the vertical gradient $\frac{\partial S}{\partial z}$ along with the flux $F_s(z_m)$ is positive (directed upward), if the concentration decreases with height ($\Delta S < 0$), and negative (directed downward), if the concentration increases with height ($\Delta S > 0$).

For parameterization of the turbulent diffusion coefficient, equations with allowance for stratification (ζ) of the atmospheric surface layer were used. For unsteadily stratified surface layer, equations (2-5) were applied.

$$K = \frac{k z_m u_*}{\varphi_h(\zeta_m)} \tag{2}$$

$$u_* = \frac{k z_m}{\varphi_m(\zeta_m)} \frac{\partial U}{\partial z} \tag{3}$$

$$\varphi_m(\zeta_m) = (1 - 16\zeta_m)^{-\frac{1}{4}}, \zeta_m < 0 \tag{4}$$

$$\varphi_h(\zeta_m) = (1 - 16\zeta_m)^{-\frac{1}{2}}, \zeta_m < 0 \tag{5}$$

where $u*$ is the friction rate [2], $k = 0.4$ is the von Karman constant, ζ is the stability parameter, $\varphi_h(\zeta_m)$ are the universal differential functions of heat and angular momentum [3] .

The stability parameter ζ_m is calculated by the iterative method with the use of the Richardson gradient number Ri by equation (6) [1]:

$$Ri = \frac{g}{\theta_0} \frac{\frac{\partial \theta}{\partial z}}{\left(\frac{\partial U}{\partial z}\right)^2} \tag{6}$$

$$Ri(\zeta_m) = \begin{cases} \frac{\zeta(0.74+0.47\zeta)}{(1+4.7\zeta)^2} & \zeta > 0 \\ 0.74\zeta\left(\frac{1-15\zeta}{1-9\zeta}\right)^{0.5} & \zeta < 0 \end{cases} \tag{7}$$

If the surface layer was steadily stratified, then ($\zeta_m > 0$), and equations (8-14) from [4] were used for the calculation.

$$K = u_* l_z \tag{8}$$

$$u_* = \sqrt{2\Psi_\tau E_z^{1/2} l_z \frac{\partial U}{\partial z}} \tag{9}$$

$$\Psi_\tau = 0.228 - 0.08 Ri_f \tag{10}$$

$$\Psi_3 = 1 - 2.25 Ri_f \tag{11}$$

$$Ri_f \approx 1.25 Ri \frac{(1+36Ri)^{1.7}}{(1+19Ri)^{2.7}} \tag{12}$$

$$l_z = z_m Ri_f \left(1 - \frac{Ri_f}{Ri_f^\infty}\right)^{4/3} \tag{13}$$

$$E_z = \left(l_z \frac{\partial U}{\partial z}\right)^2 \frac{2C_k Cr \Psi_3 \Psi_\tau}{3(1+C_r)} \left[1 - \left(\frac{3}{C_r \Psi_3} + 1\right) Ri_f\right] \tag{14}$$

where $C_k = 1.08$, $C_r = 3$, Ri_f is the Richardson flux number, E_z is the energy of fluctuations of the vertical velocity. The use of different schemes of turbulent closure, depending on the stability regimes, made it possible to obtain a full annual flux of vertical fluxes. The data filtration procedure was used to exclude errors in the obtained measurements. The gate from 5 to 95 percentile was taken for the error verification procedure. If the value was not in the gate range, it was considered an error. For the calculations, the data were divided into two periods: day-time (measurements were carried out from 13 to 17 hours) and night-time (from 1 to 5 hours). This approach allows us to estimate the fluxes in steady-state surface layer regimes.

3. RESULTS

As a result of the calculations, the values of the annual and daily dynamics of methane flows were obtained. Figure 1 shows the average long-term annual variation of day and night methane flows.

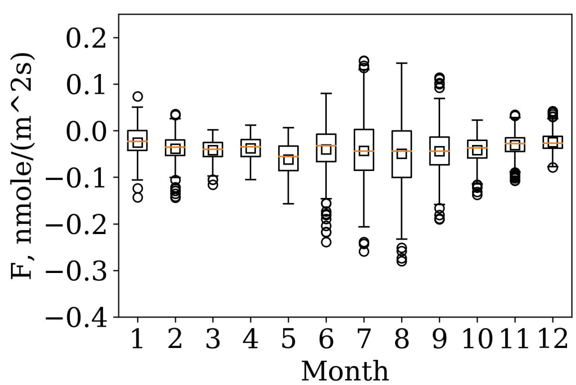

Figure 1. Annual bechvior CH_4 day time

Fig. 1 shows the negative daily fluxes in every month. The annual cycle has an extremum in the spring and summer period. The average daily methane flux values during the year vary from -0.06 to 0.025 nmol/m^{2}s and

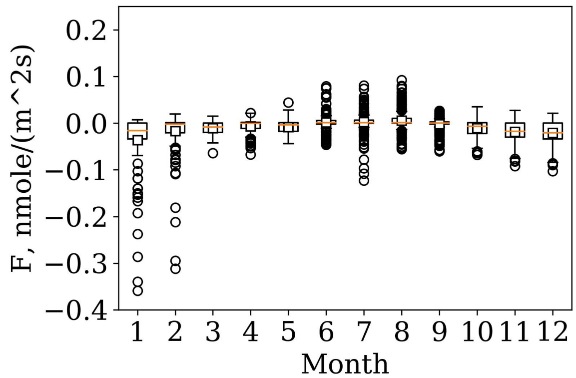

Figure 2. Annual bechvior CH_4 night time

average -0.04 nmol/m^{2}s. The median values vary from -0.055 to -0.023 nmol/m^{2}s; the average annual median value is -0.037 nmol/m^{2}s. The highest day-time flux 0.062 (-0.058) nmol/m^{2}s (starting now, in brackets are shown the median).

The annual cycle of night methane fluxes (fig.2) has two extremes: in the summer and winter periods. Between June and August, a positive value of fluxes is observed. The flux rate is higher in August. In June, the nighttime flow of methane was 1.5 (0.46) pmol/m^{2}s, and in August, 5.4 (0.97) pmol/m^{2}s. The averaged nocturnal methane flux was 3.16 (0.72) pmol/m^{2}s. The winter maximum is observed in January and amounts to -0.036 (-0.015) nmol/m^{2}s. Average night fluxes, excluding the summer months, was -0.014 (-0.011) nmol/m^{2}s. Comparing the magnitudes of the extremes of night-time fluxes in absolute magnitude for winter and summer, we can conclude that they differ by order of magnitude.

The analysis shows that methane sink in annual cycle dominates around the station. We have estimated the annual intensity of methane sink per 1 m^2. The magnitudes of the day and night sink of methane are -20.2 (-18.7) and -5.0 (-3.1) mg, respectively. The total annual methane sink is -25.2 (-21.9) mg.

4. CONCLUSION

We have concluded that the types of the annual cycle of day-time and night-time methane fluxes are different. For day-time fluxes, there is one extremum in summer. For night-time flux - two extremes; positive fluxes in the summer, and negative in another month. The presence of positive extremum of night methane flux most likely due to the activation of the natural sources (wetlands).

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