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MEASUREMENTS OF NITROGEN DIOXIDE FLUXES IN THE SURFACE LAYER FOR THE TERRITORY OF THE "FONOVAYA" OBSERVATORY

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ABSTRACT

In this paper, an assessment was made of the vertical fluxes of nitrogen dioxide using the gradient method. The calculations of nitrogen dioxide's vertical fluxes were performed by utilizing gradient measurements of nitrogen dioxide concentration and meteorological parameters at the Fonovaya Observatory of the Institute of Atmospheric Optics, Siberian Branch of the Russian Academy of Sciences from January 2017 to January 2023.

Keywords: vertical carbon dioxide fluxes, surface layer, measuring mast

1. INTRODUCTION

Measurement of nitrogen dioxide (NO2) fluxes in the surface layer of the atmosphere is an important tool for assessing the quality of the environment and public health. Nitrogen dioxide is a gaseous pollutant that is formed during fuel combustion, including from automotive vehicles [1,2]. Prolonged exposure to NO2 can cause various respiratory diseases in humans, such as asthma and chronic obstructive pulmonary disease (Special Report of the IPCC, 2019). Thus, measuring NO2 fluxes allows monitoring the level of air pollution and taking necessary measures to improve the quality of the environment. The aim of this study is to obtain an estimate of the nitrogen dioxide flux for the background area of Western Siberia, where the "Fonovaya" observatory of the Institute of Atmospheric Optics of the Siberian Branch of the Russian Academy of Sciences is located.

2. MEASUREMENT DATA AND CALCULATION METHODS

Measurements of nitrogen dioxide fluxes were carried out in the area of the "Fonovaya" [3] observatory, located in the background area of the Tomsk region (56°25'N, 84°04'E, 80 m above sea level, http://lop.iao.ru). A tall mast was installed on the observatory's territory for conducting measurements.

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
Thermo Scientific 42i	NO_2, ppb	50...1000	< 0,2 ppb	1 s

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Measurements of nitrogen dioxide concentration and meteorological parameters were conducted at two heights, 10 and 30 m. The list of equipment is presented in Table 1. The station operates in monitoring mode with a frequency of parameter registration of 1 hour. The measurements taken from January 2017 to January 2023 were used for calculating the fluxes.

To calculate the vertical fluxes of nitrogen dioxide, we utilized the gradient method based on the Monin-Obukhov similarity theory, as described in detail in [4]. The method allows for the calculation of the pollutant flux using Equation (1):

$$F_{NO_2}(z_m) = -K\frac{\partial NO_2}{\partial z} \tag{1}$$

where, $F_{NO_2}(z_m)$- is the vertical flux of nitrogen dioxide gas, K - is the coefficient of turbulent diffusion, $\frac{\partial NO_2}{\partial z}$ - is the concentration gradient, $z_m = \sqrt{z_1 z_2}$ - is the reference height for which the flux is calculated, and $z_2 = 30$ and $z_1 = 10$ are the heights of the upper and lower measurement levels, respectively. The following rule is adopted for the calculations: the vertical gradient $-\frac{\partial NO_2}{\partial z}$ and, consequently, the flux $F_{NO_2}(z_m)$, are positive (directed upwards) if the concentration decreases with height ($\Delta NO_2 < 0$) and negative (directed downwards) if the concentration increases with height ($\Delta CO_2 > 0$)).

To parameterize the coefficient of turbulent diffusion, formulas were used that take into account the stratification (ζ) of the surface layer. Formulas (2-5) were used for an unstably stratified surface layer.

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

$$u_* = kz_m\varphi_m\left(\zeta_m\right)^{-1}\frac{\partial U}{\partial z} \tag{3}$$

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

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

where, $u*$ - friction velocity [5], $\varphi_h\left(\zeta_m\right)$ - universal differential functions of heat and momentum [6], ζ - stability parameter, $k = 0.4$ - Karman's constant, $\frac{\partial U}{\partial z}$ - wind speed gradient. Stability parameter is calculated by iterative method using the Richardson gradient number "Ri" according to the formulas (6,7) [4]:

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

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

If the surface layer was stably stratified, then ($\zeta_m > 0$) and equations (8-14) from [7] were used for the calculations.

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

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

$$\Psi_3 = 1 - 2.25 Ri_f \quad (10)$$

$$\Psi_\tau = 0.228 - 0.08 Ri_f \quad (11)$$

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

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

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

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.

3. RESULTS

The conducted calculations yielded vertical flux values of nitrogen dioxide. Figure 1 presents the median values for daytime and nighttime fluxes. Daytime values were calculated for the time interval from 12 pm to 6 pm, while nighttime values were calculated for the interval from 12 am to 6 am.

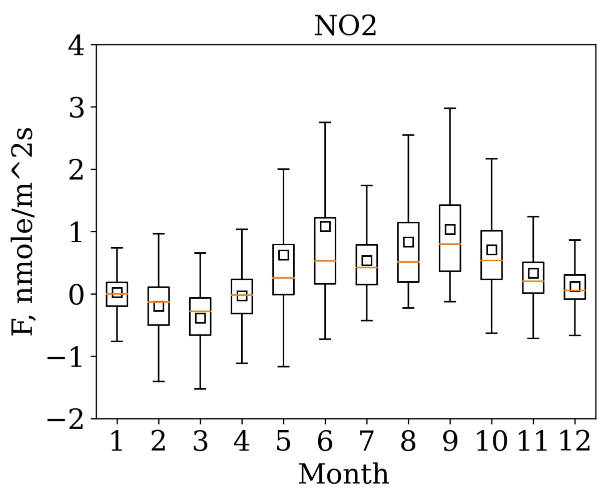

Figure 1. Monthly average values of daytime vertical nitrogen dioxide fluxes. The square indicates the mean value; the orange line represents the median; the rectangle shows the values of the 1st and 3rd quartile; the standard deviation is shown by the vertical lines.

Results depicted in Figure 1a show that the annual cycle of median daytime nitrogen dioxide flux values has two extrema. The first negative extremum in March, with a value of -0.28 nmol/(m2s), is probably due to anthropogenic pollution and transport. The second positive extremum in September, with a value of 0.8 nmol/(m2s), is caused by natural emissions of nitrogen oxides from the soil. For nighttime nitrogen dioxide flux values, natural emissions prevail. The maximum was observed in October, with a value of 0.3 nmol/(m2s). The obtained flux values are consistent with the data presented in other studies, such as [8].

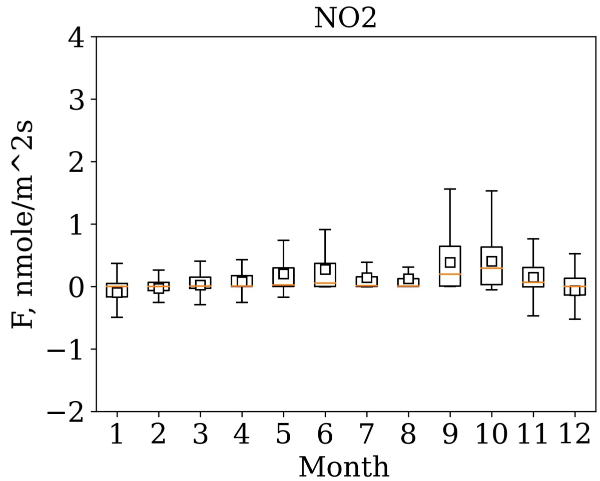

Figure 2. Monthly average values of nighttime vertical nitrogen dioxide fluxes. The square indicates the mean value; the orange line represents the median; the rectangle shows the values of the 1st and 3rd quartile; the standard deviation is shown by the vertical lines.

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