presenting author: Włodzimierz Pawlak

Net turbulent fluxes of methane and carbon dioxide in the city of Łódź, Poland – comparison of diurnal and seasonal variability

Włodzimierz Pawlak[1], Krzysztof Fortuniak[1]
[1] *Department of Meteorology and Climatology, University of Łódź,*
ul. Narutowicza 88, 90-139 Łódź, wpawlak@uni.lodz.pl

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1. Introduction

Turbulent greenhouse gases (water vapor, carbon dioxide, methane) exchange is one of the frequently undertaken research topics in urban climatology. Research on temporal and spatial variability of the turbulent fluxes of these gases are intensively realized, especially in recent years, since the relatively wide range of instruments, that enable the most accurate and suitable method for long term measurements campaign i.e. eddy covariance, become accessible (Lee et al., 2005; Foken; 2008; Aubinet et al., 2012). Carbon dioxide and methane, despite their trace presence in the atmospheric air, play a significant role in the carbon cycle (Ciais et al., 2013; Hartmann et al., 2013). They are also the greenhouse gases, which are involved in shaping of the radiation and energy balance of the Earth's surface (Ciais et al., 2013; Hartmann et al., 2013). There are natural (respiration of living organisms, methanogenesis, forest fires, cattle breeding), but also anthropogenic (fossil fuel burning, landfills, sewage and leaks from pipelines) sources of both gases (Christen, 2014). While researches on their concentration are conducted for decades (Ciais et al., 2013; Hartmann et al., 2013), their turbulent exchange between the ground and the atmosphere is still the subject of intensive, developing research. Studies on the carbon dioxide and methane amount in the air are now major priorities not only from the point of view of natural science, but also economic and social, moreover, the city is their major source as it results from the literature of the problem (Nicolini et al., 2013; Christen, 2014). Because the process of methane and other greenhouse gases exchange between the ground and overlying air is closely related to turbulent movement of air in the boundary layer, for its research the measurement techniques that allow the determination of a vertical turbulent flux of mass, energy and momentum should be used. Devices that allow the direct determination of turbulent fluxes of greenhouse gases (e.g. water vapor, carbon dioxide) were developed several years ago (half of 90s), and since then the researches were intensified. Unfortunately, the measurements of turbulent methane exchange were significantly limited due to the lack of suitable sensors, which become widely available a few years ago. It should be emphasized that the number of stations recording fluxes of carbon dioxide and methane in urban areas, is definitely insufficient in comparison to rural areas. A few hundred measurement stations were located in natural and semi-natural areas, however, measurements of carbon dioxide turbulent flux were only conducted in 20-30 cities (Grimmond et al., 2002; Nemitz et al., 2002; Kuc et al., 2003; Moriwaki and Kanda, 2004; Velasco et al. 2005; Vogt et al., 2006; Coutts et al., 2007; Vesala et al. 2008; Zimnoch et al. 2010; Pawlak et al., 2011; Gioli et al., 2012; Nordbo et al., 2012; O'Shea et al., 2012; Song and Wang, 2012; Ward et al., 2013; Christen, 2014; Crawford and Christen, 2015; Ward et al., 2015), and in case of methane this number is even smaller (Kuc et al., 2003; Zimnoch et al., 2010; Gioli et al., 2012; O'Shea et al., 2012; Nicolini et al., 2013; Christen, 2014). The problem of derivation of the diurnal and seasonal variability features of the turbulent vertical exchange of carbon dioxide, and methane especially, in urban areas and the evaluation of the weather conditions influence on the intensity of this gas exchange should be considered still open.

The measurements of mass and energy fluxes have been conducted in Łódź since autumn 2000 (Offerle et al., 2006a; 2006b; Pawlak et al, 2011; Fortuniak et al., 2013; 2015). The continuous time series of water vapor turbulent fluxes includes 15 years (with break in years 2004-2005) and 9 years for carbon dioxide (Pawlak et al., 2011; Fortuniak et al., 2013). In July 2013 the methane gas analyzer (Li7700, open-path CH4 analyzer, Li-cor, USA) was added to the measurement system.

The aim of this paper is to analyze the temporal variability and comparison to turbulent fluxes of carbon dioxide (FCO_2) and methane (FCH_4) on the basis of nearly 2-years (July 2013-April 2014) of measurements conducted in downtown of Łódź.

2. Site, measurement method and instrumentation

The measurements of FCO_2 and FCH_4 fluxes were conducted in the western part of Łódź downtown. The city is located in the Central Poland, and in the vicinity there are no orographic obstacles or large water bodies, and any large river does not flow through the city as well. Łódź is the third most populous city in Poland, the number of its population reaches approx. 706 thousand. The city center is a dense network of perpendicular urban canyons and a large number of buildings and townhouses built mainly in the nineteenth and twentieth centuries. These buildings are similar in height, have flat roofs covered with black roofing paper. In Łódź, there is a lack of characteristic for other European cities downtown with buildings clearly higher than ambient. The percentage of artificial surfaces such as buildings, streets, sidewalks, squares, etc., in the immediate vicinity of the measuring point (Fig. 1, right) is 62%. Therefore, surfaces covered with vegetation (lawns, bushes, parks, etc.) occupy 38%, with the trees, which in most cases do not exceed buildings, account for only 10% (Pawlak et al., 2011; Fortuniak et al, 2013).

For the carbon dioxide and methane turbulent fluxes measurement the eddy covariance method was applied. This method allow the determination of the flux intensity and the direction of the mass exchange between surface and atmosphere (Lee et al., 2005; Foken; 2008; Aubinet et al., 2012). According to the adopted methodology fluxes are calculated as the covariance between the fluctuations of the vertical component of the wind speed w' and fluctuations in the concentration of carbon dioxide $\rho CO_2'$ and methane $\rho CH_4'$:

$$FCO_2 = \overline{w' \cdot \rho CO_2{}'}$$

$$FCH_4 = \overline{w' \cdot \rho CH_4{}'}$$

Positive flux indicates emission while negative uptake of gas by the surface. For the determination of fluxes 1-hour averaging blocks were applied, data was averaged with simple block averaging. In computation adequate flux correction and procedure were applied: rejection the data recorded during rain, spike detection, covariance maximization, correction of air temperature measured with sonic anemometer, double coordinate system rotation (Lee et al., 2005; Foken; 2008; Aubinet et al., 2012). In addition for both fluxes the WPL correction due to changes in air density was applied (Webb et al., 1980; Lee et al., 2005; Foken; 2008; Aubinet et al., 2012). In the case of methane flux, according to the manufacturer of methane gas analyzer (Li7700, Li-cor, USA) correction terms related to air temperature, pressure and water vapor impact on spectroscopic measurement made by Li7700 sensor were included as well (Li7700 manual). Using three different tests rated stationarity of the data, the data were rejected if all three tests suggested a lack of stationarity (Fortuniak et al, 2013). In the analyzed period, 41% of the data recorded during bad weather was rejected, another 2% did not meet the stationary condition.

Fig. 1. FCO_2 and FCH_4 measurement site (left) and aerial photo of site surroundings in the western part of Łódź center (right). White lines indicate source area with probability P = 25, 50, 75 and 90% calculated for unstable conditions in the period July 2013 – April 2015. Red dotted lines indicate 250, 500, 750 and 1000 distances from the measurement site.

The measurement set-up was mounted on the mast at a height of 37 m above the ground (Fig. 1, left). The average height of the surrounding buildings was estimated at 11 m, therefore it can be assumed that the measurements were above the blending height inside the internal sublayer. During the measurements, a standard set of instruments i.e. sonic anemometer RMYoung 81000 (RMYoung, USA), infra red Li7500 CO_2/H_2O open path analyzer (Li-cor, USA) and Li7700 CH_4 open path analyzer (Li-cor, USA) was used. Fluctuations in the vertical component of the wind speed and the concentration of carbon dioxide and methane were recorded at a frequency of 10 Hz. On the basis of data collected under unstable conditions, the source area was estimated according to the Schmid methodology (Schmid, 1994; Pawlak et al., 2011; Fortuniak et al., 2013). Depending on the direction it extends from 250 to 750 meters from the measurement site.

3. Results

In the analyzed period the average carbon dioxide flux FCO_2 value was positive and reached 7.79 $\mu mol \cdot m^{-2} \cdot s^{-1}$. The average methane flux FCH_4 was also positive, but was significantly lower in magnitude, as it reached only 27.85 $nmol \cdot m^{-2} \cdot s^{-1}$ (0.0278 $\mu mol \cdot m^{-2} \cdot s^{-1}$). On average, the examined fragment of western downtown of Lodz is a net source of both carbon dioxide and methane. It can also be said that, on average, the exchange of 1 mole of methane is accompanied by exchange of 280 moles of carbon dioxide. Data shown in fig. 2 indicates that both fluxes has distinct annual variability. Due to the fact that carbon dioxide in city is of a primarily anthropogenic origin (Pawlak et al., 2011; Christen, 2014), a flux of the gas is elevated in the cold season when it exceeds 20-30 $\mu mol \cdot m^{-2} \cdot s^{-1}$. In periods of major coolings its value reaches nearly 60 $\mu mol \cdot m^{-2} \cdot s^{-1}$. Elevated winter FCO_2 values are the results of increased fossil fuels combustion (home heating and cooking, car engines). Summer values are significantly lower and rarely exceed 20 $\mu mol \cdot m^{-2} \cdot s^{-1}$. In the summer traffic is less intense (holidays) and the combustion of fossil fuels in houses that do not require heat also is severely limited. Another reason for the reduced value of FCO_2 and even the appearance of negative flux is biological activity of urban vegetation. During photosynthesis, plants absorb carbon dioxide from the air to some extent offsetting anthropogenic emissions. The methane flux (Fig. 2) has a similar variation with higher values in winter (up to 200 $nmol \cdot m^{-2} \cdot s^{-1}$ and more) , and reduced in the summer (up to 100 $nmol \cdot m^{-2} \cdot s^{-1}$). The reason for such variability can probably be the burning of natural gas, and leaks from pipelines that are higher in the cold season when gas consumption is higher (Lowry et al., 2001; Nam et al., 2004; Wennberg et al., 2012; Phillips et al., 2013).

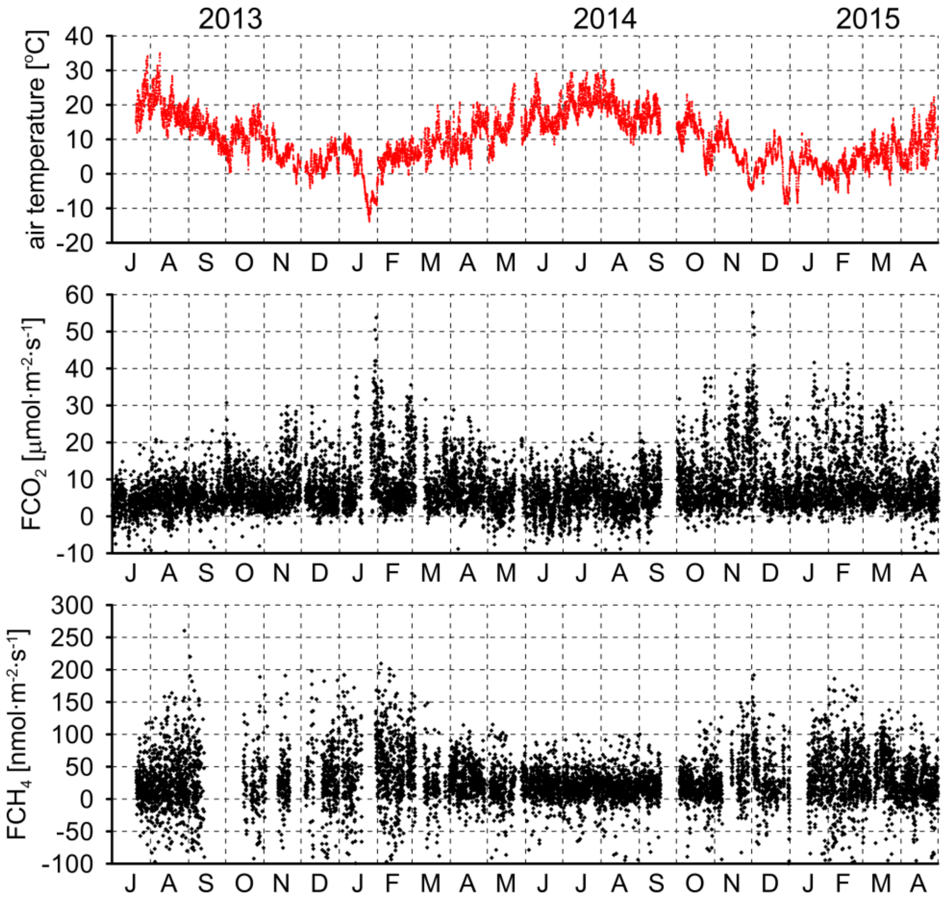

Fig. 2. One hour means of air temperature, carbon dioxide FCO_2 and methane FCH_4 turbulent net fluxes in the center of Łódź in the period July 2013 – April 2015.

Turbulent fluxes of carbon dioxide and methane are also characterized by a diurnal rhythm (fig. 3, left and the middle) with higher values during the day and lower in nighttime. On average, however, they have positive values which indicate that the tested part of the city is a source of carbon dioxide and methane throughout the day. In both cases, increased daily values are a result of diurnal rhythm of turbulence. In the diurnal cycle of FCO_2 flux (fig. 3, middle, red line) there are two peaks: one occurring in the morning and the second in afternoon-evening. They reflect morning and afternoon peak in traffic and afternoon house residents activities (cooking, heating, etc.). The daily variability of the methane flux is not as pronounced as in the case FCO_2 (fig. 3, left, red line). Except for elevated diurnal values it is hard to find the similar values to FCO_2 (fig. 3, middle, red line).

Since both studied gases are produced during the anthropogenic processes, variability of FCO_2 and FCH4 fluxes has also clear weekly rhythm (fig. 3). It is particularly evident in the case of the carbon dioxide flux, which is significantly higher during working days (FCO_2 is increased by 1-2 $\mu mol \cdot m^{-2} \cdot s^{-1}$), and lower (by 2-3 $\mu mol \cdot m^{-2} \cdot s^{-1}$) during the weekends. In the case of the methane flux, the situation is similar - FCH_4 on working days is increased by approximately 2-3 $nmol \cdot m^{-2} \cdot s^{-1}$, whereas at the weekend observed values are reduced by approximately 3-5 $nmol \cdot m^{-2} \cdot s^{-1}$. At 12 am and 8 pm the values observed during the weekend exceed those recorded during the working days, what is hard to explain.

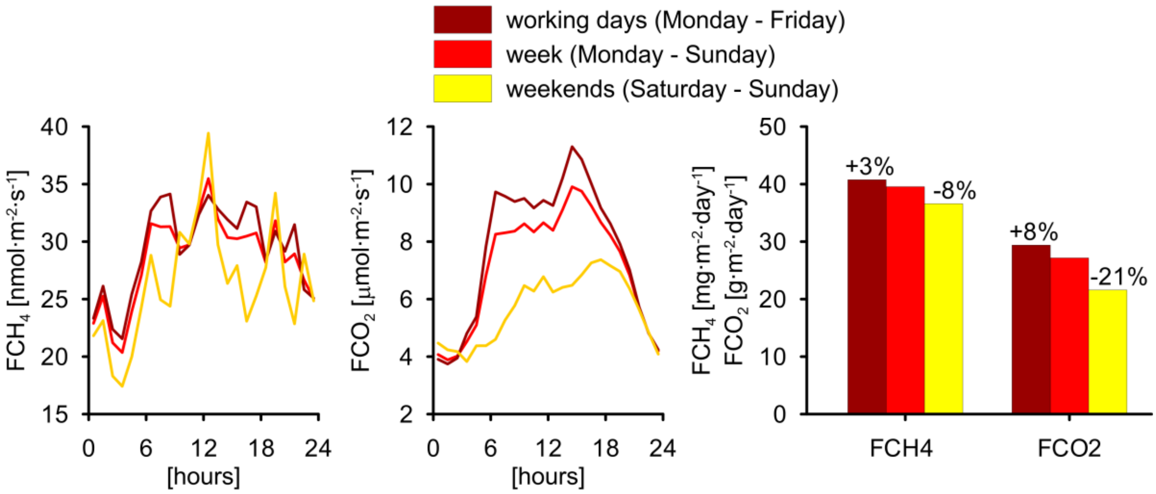

Fig. 3. Mean diurnal variability of methane (left) and carbon dioxide (middle) net turbulent fluxes and mean daily turbulent exchange of methane and carbon dioxide (right) in the center of Łódź in the period July 2013 – April 2015 calculated for weeks (red line) and separately for working days (brown line) and weekends (yellow lines).

On the basis of diurnal variations of FCO_2 and FCH_4 the average daily exchange of carbon dioxide and methane for the whole week and for weekdays and weekend days were determined. In both cases, during the working days, the western part of Łódź center emitted elevated amount of carbon dioxide and methane, and lower on Saturday and Sunday (fig. 3, right). On average, during the working day, this part of city emitted 8% more than the average, while during the weekend 21% less than the average. In the case of methane contrast was smaller, the values were respectively +%3 and -8%.

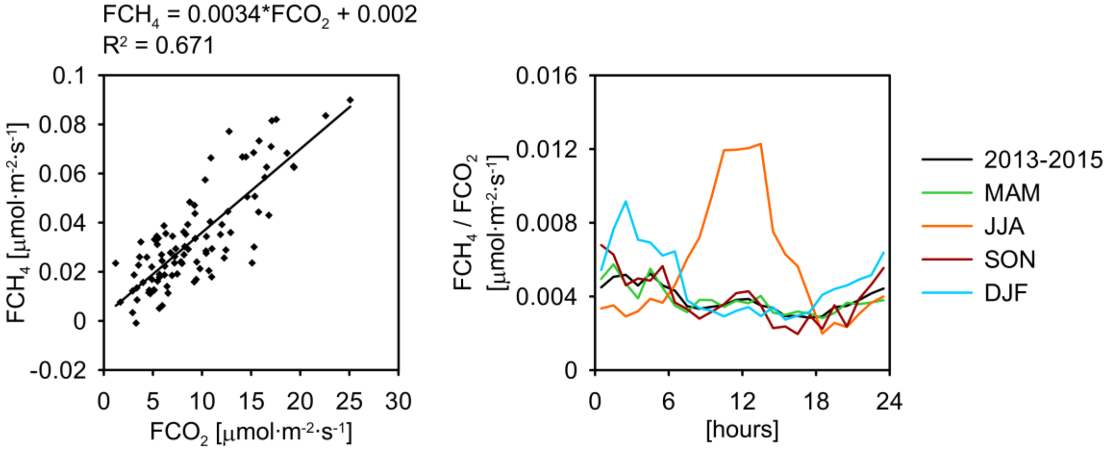

Fig. 4. Mean daily turbulent fluxes of methane in relation with mean daily fluxes of carbon dioxide (left) and mean diurnal ratio of methane to carbon dioxide fluxes (right) in the center of Łódź in the period July 2013 – April 2015.

Comparison of mean daily values of carbon dioxide and methane fluxes revealed a correlation (fig. 4, left - for the computation only the days, when at least 20 of the 24 1-hour blocks of FCO_2 and FCH_4 were recorded). FCO_2 increase is accompanied by an increase in FCH_4, with a coefficient of determination of the order of 0.671. The diurnal variability of FCH_4/FCO_2 ratio was estimated for the whole measurement campaign and seasons of the year (fig. 4, right). In the daytime the value of this coefficient is approximately constant and reaches up approx. 0.004. The exception is the summer when the value of the coefficient is about 3 times greater.

4. Summary

The measurements indicate the existence of similarities between the temporal variability FCO_2 and FCH_4 fluxes. The methane flux has, of course, far lower values, due to about 200-fold lower concentration of this gas in the air. Both fluxes have a similar features of annual and diurnal variation. Similar measurements and comparisons carried out in other cities could give an answer to the question whether the FCO_2 flux can be used to estimate the FCH_4 flux.

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