Author reply: Eddy covariance measurements of the net turbulent methane flux in the city centre – results of 2 year campaign in Łódź, Poland by W. Pawlak and K. Fortuniak

Referee 1 comments in bold

We would like to thank referee for his thorough reading of the manuscript and for very detailed, constructive and useful comments, which show his dedication to improving this manuscript.

This is a new and important study, in which a long time-series (2 years) of methane fluxes is presented and analysed. The dataset represents the first of its kind in terms of duration, enabling annual total methane flux to be estimated for an urban area, as well as some consideration of inter-annual variability. The analysis focuses on the temporal patterns in the methane flux at various timescales (daily, weekly, monthly, seasonally and annually) and spatial patterns are explored too. In some places deeper analysis may offer additional insight.
The article is generally well written and thorough, but very repetitive in places. Sometimes use of unnecessary words obscures the meaning. I have made some specific suggestions below but **the article as a whole would benefit from being more concise. Shorter (more) paragraphs would also improve readability. Overall the quality of the work is high and I recommend this paper for publication in ACP following minor revisions.**

Specific comments:

1. Introduction: The introduction would be easier to read if it was more concise and less repetitive. Breaking the text up into smaller paragraphs would also help. I have made some suggestions below.
According to referee comments this section has been rewritten and divide to smaller paragraphs

Introduction:
"The temporal and spatial variability of greenhouse gas fluxes in the atmosphere is at present one of the most widely discussed climatological problems. Methane, despite its trace presence in the air (ca 1.8 ppm, Hartman et al., 2013), plays an important role in the environment. It participates in the global carbon cycle and is considered one of the greenhouse gases whose concentration in the atmosphere affects the radiation balance of the Earth's surface. An increase in the concentration of methane contributes to an enhancement of the greenhouse effect; therefore, the emissions of this gas to the atmosphere should be carefully monitored.
Methane is produced during the process of methanogenesis under anaerobic conditions, from the decay of organic plant debris in water. The most important source of methane in the world is wetlands (Shurpali et al., 1998; Rinne et al., 2007; Baldocchi et al., 2012; Hatalaa eta al., 2012), but also paddy fields (Miyata et al., 2000), cattle farming (Laubach and Kelliher, 2005; Dengel et al., 2011; Hartmann et al., 2013; Nicollini et al., 2013), as well as emissions from the soil (Smeets et al., 2009; Denmead et al., 2010; Wang et al., 2013). Moreover, emissions of methane accompany forest fires and grass vegetation. The effect of the combustion of natural gas (which contains at least 80% methane) is mainly water vapour and carbon dioxide. The combustion of fossil fuels is, however, predominantly incomplete, and therefore it is an important factor causing anthropogenic methane emissions. This happens in the case of combustion of both natural gas and hydrocarbons contained in petrol and other fuels (Nam, 2004; Nakagawa et al., 2005; Wennberg et al., 2012). Another important source of methane in urbanized areas is leakage from urban gas pipelines (Lowry, et al., 2001; Gioli et al., 2012; Wennberg et al., 2012; Phillips et al., 2013). Methane may also be emitted during the anaerobic respiration of bacteria in urban soils (Bogner and Matthews, 2003) and in the course of decomposition of solid waste and wastewater in sewage systems and at landfill sites (Bogner and

Matthews, 2003; Laurila et al., 2005; Lohila etal., 2007; Wennberg et al., 2012; Jha etal., 2014). On the other hand, certain soil bacteria consume methane, which is one of the processes of its removal from the air (Goldman et al., 1995; Kaye et al., 2004; Groffman et al., 2006; Groffman and Pouyat, 2009). Methane is involved in some of the reactions leading to photochemical smog formation (Seinfeld and Pandis, 2006). The disintegration of methane also results from its reacting with the hydroxyl group in the atmosphere (Whalen, 2005). Annual global emission of methane to the atmosphere has been estimated as ~5000 Tg of CH4, and emission from landfills and waste (87-94 TG of CH4) or fossil fuels (85-105 Tg of CH4) are 2-3 times lower than estimated emission form wetlands (177-284 Tg of CH4) (Ciais et al, 2013).

Research on the methane content in the air is now a priority because, as it follows from the literature on the problem, the city may be a significant source of this gas (Elliot et al., 2000; Gioli et al. 2012; O'Shea et al., 2012; Nicolini et al., 2013; Phillips et al., 2013; Christen, 2014; Kumar and Sharma, 2014). The measurements of changes in CH4 concentrations have been carried out for decades (Ciais et al., 2013; Hartmann et al., 2013), while the analyses of its flux, especially in urban areas, are extremely rare. In recent years, there have been approximately 500 stations measuring the fluxes of CO2 around the world, of which only ca 20 are located in cities and only a few were able to measure methane flux (Nordbo et al., 2012; Oliphant, 2012; Christen, 2014). It can be concluded that the measurements of methane fluxes in the cities are in the early stage and there are still some challenges like long term measurements (much longer than a few weeks or months) and relationship between methane fluxes and land use.

Basics of theory and measurement techniques of turbulent exchange of mass, energy and momentum fluxes have been developed for decades (Stull, 1988; Lee et al., 2005; Foken, 2008; Aubinet et al., 2013). The measurements of the fluxes of methane were severely limited due to the lack of suitable sensors which to have appeared a few years ago (Pattey et al. 2006; Hendricks et al., 2008; Eugster and Pluss, 2010; Dengel et al., 2011; Detto et al., 2011; Sakabe et al., 2012). At present, the most widely used instrument is in all probability the LI7700 Open Path CH4 Analyzer (Burba and Anderson, 2010; McDermitt et al., 2011) and eddy-covariance as a measurement technique (Aubinet et al., 2012). All over the globe, there are only a few long-term, continuous measurement series of turbulent fluxes of water vapour and carbon dioxide recorded in urban areas (Christen, 2014). In the case of methane flux, such series are probably at the implementation phase, since previous studies focused on areas which are the largest source of methane, i.e. natural wetlands (Shurpali et al., 1998; Rinne et al., 2007; Baldocchi eta al., 2012; Hatalaa eta al., 2012; Aubinet et al., 2013), agricultural land (paddy fields, Miyata et al., 2000) or over forests (Smeets et al., 2009; Wang et al., 2013). The chamber method, widely used in rural areas, has only a limited relevance in the city: it makes it possible to take measurements of methane emissions from the specific areas like urban lawns (Baciu et al., 2008), however, it cannot be used in the case of larger urban areas. A variety of techniques have recently been applied to provide independent estimates of urban CH4 emissions like airborne observations (O'Shea et al., 2014; Mays et al., 2009), Fourier Transform Spectrometry (Wunch et al., 2009) or isotopic source apportionment studies (Lowry et al., 2001). Morizumi (1996), in turn, suggested the occurrence of covariability of radon Rn-222 and the methane flux concentrations, which, based on this, he estimated to be 20 mg·m-2·day-1. In Poland, the issue of exchange of greenhouse gases in an urban area is studied, besides Łódź, in Cracow where, based on the measurements of CH4 concentrations and the height of the atmospheric boundary layer, the average monthly nocturnal flux of methane has been estimated to be 0.8 do 3 mg·m-2·h-1 (Kuc et al., 2003; Zimnoch et al., 2010).

The aim of this study is to analyze the temporal variability of the turbulent flux of methane (FCH4) based on a long-term series of measurements recorded for over two years in the centre of Łódź between July 2013 and August 2015. The diurnal variability of FCH4 was analysed and monthly values of the flux were determined and an attempt was undertaken to assess the cumulative annual exchange of methane between an urban area and the troposphere in order to determine whether the centre of Łódź was an equally efficient source of methane to the troposphere as of carbon dioxide. The measurement results were compared to the variability of selected meteorological elements. As *the methane emissions in the city are determined mainly by anthropogenic factors, the values of fluxes on weekdays and at weekends were compared. Due to the impossibility to obtain relevant data, there was no comparison made with the values of fluxes using specific inventory methods. "*

P2 L15-18: To improve readability delete unnecessary words: 'In the first place', 'furthermore', 'the formation of'
Done

P2 L27-8: Delete ', and methane is also the main component of natural gas' as it is unnecessary given L28-9.
Done

P3 L24 – P4 L8: This part is quite hard to follow, a bit repetitive and could benefit from rewording.
According to reviewer comments this section has been rewritten.and now reads:
*"*Unfortunately, the lack of precise fast response instruments resulted in the fact that initially it was only possible to use indirect methods for fluxes measurements, such as the gradient method or the chamber method (Nicolini et al., 2013). The instruments to measure the turbulent fluxes of greenhouse gases such as water vapour and carbon dioxide became more widely available several years ago and since that time (the first half of the 1990s) research are significantly intense (Aubinet et al. 2012). The measurements of the turbulent exchange of methane were severely limited due to the lack of suitable sensors which to have appeared a few years ago (Pattey et al. 2006; Hendricks et al., 2008; Eugster and Pluss, 2010; Dengel et al., 2011; Detto et al., 2011; Sakabe et al., 2012). At present, the most widely used instrument is in all probability the LI7700 Open Path CH_4 Analyzer (Burba and Anderson, 2010; McDermitt et al., 2011). The number of publications describing the results of measurements of the turbulent flux of methane is therefore relatively small, in contrast to the turbulent process of carbon dioxide exchange, although it should be noted that this kind of research in urbanized areas is still relatively rare (Nordbo et al., 2012; Oliphant, 2012). In recent years, there have been approximately 500 stations measuring the turbulent exchange of CO_2 around the world, of which only ca 20 are located in cities (Nordbo et al., 2012; Oliphant, 2012; Christen, 2014)."

P4 L20-22: This sentence is not clear, please rephrase.
This sentence has been rewritten: "The chamber method, widely used in rural areas, has only a limited relevance in the city: it makes it possible to take measurements of methane emissions from the specific areas like urban lawns (Baciu et al., 2008), however, it cannot be used in the case of larger urban areas."

P5 L18: Delete 'Furthermore'
Done

P5 L19: Delete 'in the following months'
Done

2.1 Study area and site location: This section is long and quite hard to follow, in particular there are many details given about various parts of the city and it does not convey a clear message to the reader. It may be easier to follow if the section was restructured, so that after the description of the city (P5 L30 – P6 L8) comes the description of the measurement location (P6 L28 – P7 L9) starting with 'The measurements of

methane: : : persons per km2' (P7 L8-10) and then the land cover description (P6 L10 – L27). This should make it easier to identify exactly which areas are being discussed in relation to the city as a whole, as opposed to the measurement location and source area. Please also reduce repetition and unnecessary text.

According the reviewer comment this section has been reconstructed. Moreover we agree that some details are not necessary and has been removed from the text:

"Łódź is one of the largest cities in Poland. The area of the city is about 295 km2, and its population is estimated at about 706 thousand residents. The city is located in central Poland, on relatively flat terrain sloping south-westwards (its altitude varying from ~280 to ~160 m.a.s.l.). The most densely built-up city centre covers an area of 80 km2 and the altitude differences in this part of town do not exceed 60 m. In the immediate vicinity of Łódź, there are no large bodies of water, rivers or orographic obstacles, which facilitates investigating the climate of the city. Another factor making it easier to take measurements of turbulent fluxes of mass and energy in Łódź is that the city, unlike other large cities in Poland, does not have a standard central sector of tall buildings, clearly towering over the urban canopy layer.

The measurements of turbulent fluxes of methane are conducted in the western part of the city centre (51°47'N, 19°28'E, Fig. 1), in an area with the highest population density, reaching 17.2 thousand persons per km2. The station for measurements of fluxes of mass, energy and momentum has been operating in the western part of Łódź since 2000 (Offerle et al., 2006a; 2006b, Pawlak et al., 2011; Fortuniak et al., 2013, Fortuniak and Pawlak, 2014), but methane fluxes have been studied since July 2013. The measurement set is mounted on top of a mast at a height of z = 37 m (Fig. 2, left), which, given the average height of buildings of 11 m, enables the assumption that the measurements are taken above the blending height in the inertial sub-layer (Fig. 2). The source area of turbulent fluxes was estimated (Fig. 1) for data collected during unstable stratification ((z-zd)/L < -0.05) around midday (10.00 – 14.00) following the method of Schmid (1994) and ranged from 250 to 750 m away from the measurement station (Fig. 1).

The percentage of artificial surfaces (buildings, sidewalks, streets, squares, etc.) in this part of town is 62%, the remaining part being covered with vegetation, of which only 10% are trees (Kłysik, 1998). The vegetation is distributed unevenly in the form of lawns and trees growing along the street canyons. In the immediate vicinity of the measurement point, 3-5 storey 15-20 m high buildings dominate, built mostly in the 20th century. Most of them are characterized by flat roofs covered with black tar paper or sheet metal. The trees growing in the area are mostly deciduous and their height usually does not exceed the height of the buildings, which results in a well-formed roof surface with an average height of 11 m. The density of built-up areas north and east of the measurement point as compared to the southern and western sectors is 10-20% greater (Fig. 1). The displacement height zd is estimated at 7.7 m. According to the classification by Stewart and Oke (2012), the local climate zone can be described as compact low rise. The roughness coefficient z0m estimated for the neutral stratification surrounding the measurement point was on average ~2.5 m. More information on the city's structure and the local climate conditions can be found, e.g. in Kłysik (1996), Kłysik and Fortuniak (1999), Fortuniak et al. (2006, 2013), Offerle, (2006a, 2006b), Pawlak et al. (2011) and Zieliński et al. (2013). The gas distribution network and sewerage system around the flux tower are shown in Fig. 1."

P6 L4: Delete 'definitely'

Done

P6 L8: Start a new paragraph at 'The measurements: : :'

Done

P6 L10-11: It is not clear what is being 'averaged' here. May be best to omit 'average' and change 'reaches' to 'is' or provide further details (e.g. mention wind sectors used).

Done

Now this sentence reads:
"The percentage of artificial surfaces (buildings, sidewalks, streets, squares, etc.) in this part of town is 62%, the remaining part being covered with vegetation, of which only 10% are trees (Kłysik, 1998)."

P6 L18-20: May be best to switch these two sentences describing the general study area with the previous one which talks about differences between sectors.
Done

P6 L14, L20, L22: There are several different heights given here, presumably for different areas of the city. However, it is not clear to the reader which are the relevant heights, i.e. those that are considered to be within the measurement footprint and used to estimate the roughness and displacement height. Are the '10-12 storey buildings' within the footprint? Have they been included in the calculation of z0, but not zd?
No, these buildings are quite far away (3-4 km from the site location) and definitely outside the footprint. They haven't been included in the calculation both z0 and zd. It was just an information about the other districts of the city but we agree that this is not necessary. The sentence "The centre is surrounded by industrial and residential areas with tall 10-12 storey buildings or loosely built-up with single-family houses." has been removed from the manuscript.

The values given here seem to match those given in Pawlak et al. (2011), so it may be worth directing the reader to that reference in particular. However, different values are given in Offerle et al. (2006a). Please explain.
After Offerle's and ours research in the years 2000-2003 we were able to obtain a more accurate database of buildings height. Urban canopy layer height has been recalculated and final values of zH and zd are a little bit different than in Offerle (2006a).

It would also be useful to give the site name so that readers can relate this work easily to previous work carried out at the same location.
We are not sure if the giving a name of station would be appropriate. In some of the publications listed in the lines P6 L26-27 concerning the center of Lodz " Lipowa station" is used but not in others.

P6 L31-33: To avoid repetition delete this part ('when : : : air') as it effectively says the same as the first half of the sentence and this point has already been made earlier in the paper as well.
Done

P7 L3-7: Suggest rephrasing as, 'The source area of turbulent fluxes was estimated (Fig. 1) for data collected during unstable stratification ((z-zd)/L < -0.05) around midday (10.00 – 14.00) following the method of Schmid (1994).'
Done

P7 L7-9: Could rephrase as, 'The source area ranged from 250 to 750 m away from the measurement station.'
Done, the sentence now reads:
"The source area of turbulent fluxes was estimated (Fig. 1) for data collected during unstable stratification ($(z-z_d)/L < -0.05$) around midday (10.00 – 14.00) following the method of Schmid (1994) and ranged from 250 to 750 m away from the measurement station (Fig. 1)."

P7 L9-17: This part does not really belong in the site description – it would fit better in the results. Suggest simply saying 'The gas distribution network and sewerage system around the flux tower are shown in Fig. 1' here, and saving the methane discussion for the results. (Note Fig. 2 in L17 should be Fig. 1.)

Done

2.2 Instrumentation and data processing: This section is generally clear but splitting into smaller paragraphs would help improve readability. One or two sentences may need to be moved around so that each paragraph deals with a particular topic before moving on to the next. P7 L20-30: Suggest rewriting more concisely.
Regarding to referee comment this section has been divided on smaller paragraphs but without giving them separate titles. The initial part of the section (P7 L20-30) has been rewritten and now reads: "The measurements of the turbulent fluxes of methane were carried out using a standard measurement set consisting of an ultrasonic anemometer RMYoung model 81000 (RMYoung, Traverse City, Michigan, USA) and a methane fluctuation sensor with an open measurement path LI7700 (Li-cor, Lincoln, Nebraska, USA). Due to the fact that the final calculation of methane flux also requires the values of sensible heat and water vapour fluxes in the place of observation (LI 7700 instruction manual), the measurement set also included a sensor of the fluctuations of water vapour and carbon dioxide LI7500 Infra Red CO2/H2O open path analyzer (Li-cor, Lincoln, Nebraska, USA)."

P8 L21: Delete 'during the measurements'
Done

P8 L23, L26: Better to use 'RSSI' throughout this sentence, as opposed to switching between RSSI and signal strength of the instrument/signal strength value.
Done

P8 L21-26 and Fig. 2: Mention that Fig. 2 indicates the cleaned dataset (RSSI > 20%), e.g. in the figure caption.
Done, now caption reads:
"Figure 2. FCH4 measurement site in Łódź (left) and instrumentation (middle). Right figure show frequency of measured 1-hour blocks of raw data in relation with RSSI (Received Signal Strength Indicator) of Li7700 methane open path analyzer. Data recorded only in the case RSSI>20% were taken into account."

This should also be made clearer for the percentages discussed in the text, e.g. ': : :RSSI > 20% were chosen. Of these, RSSI exceeded 70% in only about 8% of cases: : :'
Done

P9 L10: Delete 'In the calculations'
Done

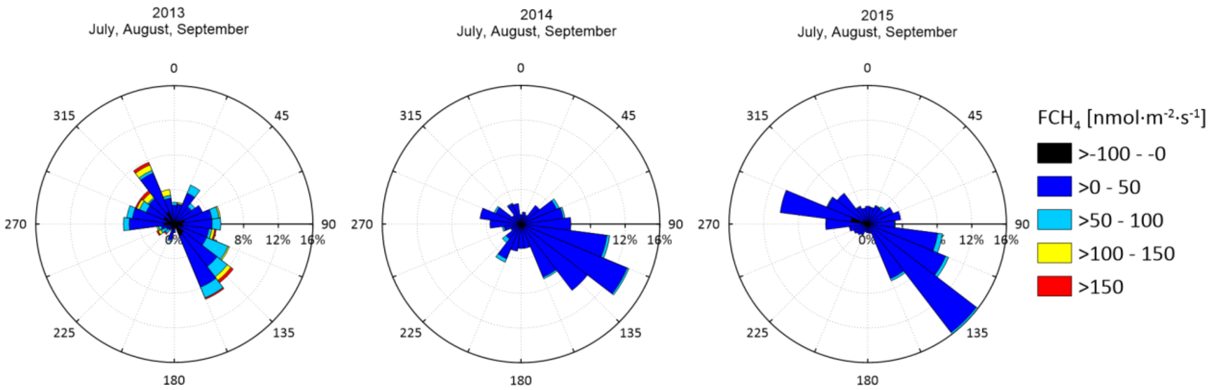
P10 L2: Delete 'earlier'
Done

3. Results: Suggest new paragraphs at P10 L28 (stability), P11 L8 (fluxes).
Done

P11 L32: New paragraph before 'It seems: : :'
Done

P11 L31-32, P12 L25: Could the authors suggest an explanation for the exception in July and August 2013? A little more discussion would be helpful (e.g. compare bottom two panels in Fig 3 and the statistics in Table 2 – the variability seems to be possibly more of an exception than the average values)

Indeed July, August and September 2013 are a surprising exception in the series. On the basis of the statistics in the Table 2 it can be concluded that the differences are very significant in the case of averages (vs. 2014 and 2015), but the median differ are significantly less. It may suggest some "incidents" with increased emission of methane in the surroundings of site in summer 2013. Unfortunately it is very difficult for us to give the reasons for this phenomenon. In the period July-September 2013, 2014 and 2015 no difference in the directions of airflow has been found and E-SE and W-NW dominates (see attached figure). In 2013, however, there were significantly higher values, which could affect the averages. Elevated FCH4 fluxes in the E-SE may be associated with the renovation of which at the time was implemented (construction of a tunnel for one of the main streets in the city - main E-W oriented street on the FIG. 1). During the implementation of the investment deep excavations were made and it is possible that some leakage or unsealing of gas pipelines could appear. Unfortunately, we do not have any data to prove it, so this is just our hypothesis (investment has been finished in autumn 2013). In turn, we don't have any ideas about the reasons of elevated FCH4 fluxes from NW sector in 2013.



Information about it has been added to the text and reads:
"The exception was the summer of 2013 when in July and August the recorded values of FCH4 were close to winter values. However, only average values were elevated, while the median values are similar to those of July and August 2014 and 2015. It can be assumed that in the summer of 2013 additional sources of methane occurred, which could be the result of damages of the gas network on SE form the station where the deep excavations associated with the construction of a tunnel for one of the main streets of the city center has been done (see main EW oriented street from the Fig. 2)."

In Fig 4 months in different years have been grouped together to create the daily cycles. Are the patterns significantly different if the years are considered separately (in general and particularly for summer 2013)?
There are no significant differences except summer 2013 months

P12 L4: Is there any evidence for increased discharge from motor vehicles in winter? Is this related to combustion conditions or the traffic load?
No we don't have such evidence. This issue raises our doubts and we regret that we cannot explain it on the basis of actual data. As we mentioned this in the manuscript, unfortunately, we do not have access to any inventory data Including traffic load. On the basis of years of observation can be noted, however, that in the winter, traffic is in Lodz less organised and regular. Definitely higher than during other seasons is the number of traffic jams in which the cars are standing or very slowly moving and fossil fuels combustion is then much more extremely intense than normal motion when the cars go smoothly at a speed of 50-70 km/h.

P12 L30: Suggest deleting 'Most importantly' – unless the annual variability presented in Section 3.2 is in doubt.

Done

The description of time needs to be more precise so that the reader understands exactly what is meant. Examples include P13 L4: 19.00-20.00 is probably too late to call 'afternoon', 'evening' is better. P13 L6: Change 'during the noon hours' to 'around noon', 'around midday' or 'during the middle of the day'. P13 L9: Does 'before noon' refer to the morning maximum (7.00-8.00)? Might be better to say 'in the morning'. P13 L11: Talks about the increased distribution 'during the day' but FCH4 is lowest during the day and peaks in the morning and evening.
Description of time in this section has been clarified.

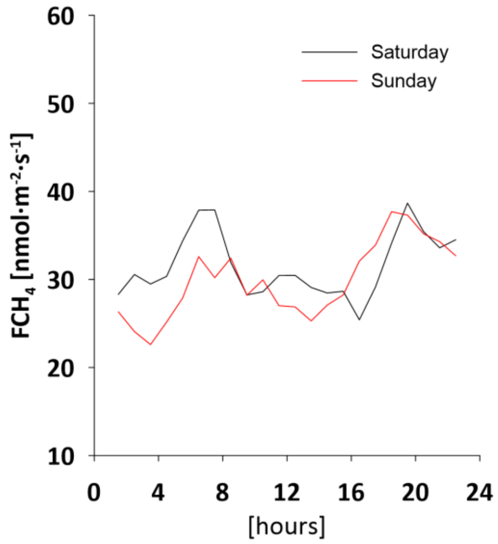
Fig. 5 and Fig. 6 are excellent. The discussion accompanying Fig. 6 is very clear.

P15 L24-26: As the partitioning of the sources of the observed methane flux is unknown, it is not possible to say that it is the anthropogenic sources that are less active at weekends. Rather, the weekday/weekend comparison suggests that the sources are likely to be anthropogenic because the observations show weekday/weekend differences.
We agree with referee's opinion. The sentence has been rewritten and now reads:
"These results suggest that, on average, in the study the period anthropogenic sources of methane are likely to be less active at weekends as compared with working days."

P15 L26-P16 L4: These differences are small and the inconsistency between summer/ winter and spring/autumn makes it hard to draw clear conclusions (as indicated by the authors). Further analysis and discussion may be informative. If the data are examined by month do the same seasonal differences emerge, or is this behaviour due to particular months skewing the results?
We have decided to analyse the differences in working days / weekends for the whole period and for seasons only. The reason is that after the data quality control, dataset has been shortened (Table 1) and we were worried that calculations for shorter time periods may introduce inaccuracy and unnecessarily complicate analysis.

Are the findings any clearer if holidays are taken into account as well as Monday-Friday and Saturday-Sunday differences?
No there are no significant differences

Are there significantly different fluxes on Saturday compared to Sunday, reflective of people's behaviour?
Regarding to referee comment we have compared Saturday and Sunday fluxes and there are some differences. Both on Saturday and Sunday, there are two peaks in the diurnal course (morning and evening) and the overall fluxes are lower in comparison with the working days. On Saturday, the average flux is, however, lower by 6.9%, while on Sunday by 12% thus almost doubled. The difference is a result of significantly lower peak on Sunday morning (see attached figure), which can be attributed to a typical human activity on Sunday- people sleep longer, later use natural gas for cooking, traffic load is also much less than at the same time of day on Saturday.



Information about this has been added to the text and reads:
"These results suggest that, on average, in the study the period anthropogenic sources of methane are likely to be less active at weekends as compared with working days. It should be emphasized that on Saturday, the average flux was lower by 6.9% in comparison with working days one, while on Sunday by 12%. The difference is a result of significantly lower peak on Sunday morning, which can be attributed to a typical not intensive human behaviour on Sunday morning and lower traffic load in comparison with the same time of day on Saturday."

Are there temperature differences between weekdays and weekends which may explain some of the trends seen?
No, there are no temperature differences

The wind direction analysis is interesting but would benefit from deeper examination and discussion. The large vegetated area to the west of the flux tower should be mentioned. Observed FCH4 also appears to be lower for southerly wind directions, potentially coinciding with the undeveloped area and scarcity of gas and sewerage pipes (Fig. 1). Is the large intersection to the south-west likely to have heavier traffic loads compared to other roads in the area?
Done, information about FCH4 from the different sectors has been added to the text: "As mentioned in Section 2.1, building density in the centre of Łódź is the most densely built-up area of the city. The measurement point is located in an area of uniform similar building density parameters, while, as mentioned in section 2.1, this density is slightly greater to the east and north of the station. An analysis of the average value of FCH4 depending on the wind direction confirms, at least in part, the impact of building density on the value of methane turbulent exchange (Fig. 8). The fluxes of CH4 recorded during airflow from the north, and especially from the south-east, were by far the largest in the study period and reached 35-45 nmol·m-2·s-1 (Fig. 8, left). However, it is difficult to regard the relationship urban design-FCH4 as certain, due to the increased values of FCH4 coming also from the south-western sector (approximately 40 nmol·m-2·s-1). Of course, such a relationship cannot be ruled out; however, the local point sources of methane may play an important role, but are difficult to identify. In the case of the south-western sector, the LPG station located approximately 800 m from the measurement station may be such a source, like lying about 200 m further to the west large intersection where traffic load is usually larger than in the surrounding streets. Significantly lower values of FCH4 (less than 20 nmol·m-2·s-1) observed during the airflow from the south and west may be due to the presence of large urban parks (Fig. 1). Through these areas, first of all, do not run the

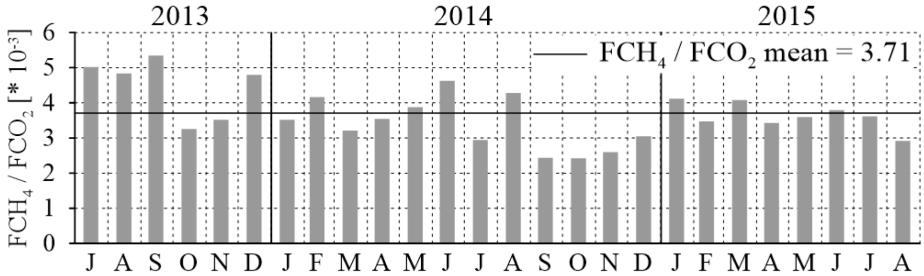
streets with car traffic, and on the other hand, the density of the gas network and sewage system is also significantly smaller in comparison with other sectors (Fig. 1).”

The discussion needs to explore further than building density – i.e. the results should be related to the networks shown in Fig. 1. More could be made of the temporal differences in the spatial patterns (i.e. the similarity in the magnitude of FCH4 observed during both warm and cold seasons to the west, versus major changes in other wind sectors). If sufficient data are available, the authors may want to consider generating seasonal diurnal cycles for different wind sectors (or groups of wind sectors) to explore whether temporal signatures typical of anthropogenic behaviour (rush hours, weekday/weekend differences, seasonal heating demand) offer additional insight into the various CH4 sources.
As it was written in section 2 detailed quality control and rejection of the data recorded during precipitation has led to a substantial reduction in the number of data suitable for analysis. Such attitude aimed to provide assurance that the analysis of FCH4 fluxes will be carried for only the correct high quality data, but, on the other hand, significantly reduced the data series. For this reason, we have decided to reduce analysis of FCH4/wind direction relation to the basic information.

P16 L18: Could the LPG station be responsible for the increased FCH4 observed over the broad range of south-westerly wind directions? Does it coincide with the 230-240_ wind sector during the warm period? It may be helpful to mark the LPG station (and any other identifiable point sources) on Fig. 1. What about the high fluxes seen between 70-80_ in the cold period?
Identification of methane emission sources without the inventory data is very difficult. Moreover there were also no possibility to perform additional concentration of methane measurements in places with, probably, increased emissions (e.g. local sources like mentioned gas station). LPG station, as previously mentioned area of road tunnel construction are our suspicions about potential sources of methane in the vicinity of the measuring station. On Figure 1 both places has been marked.

Findings from the earlier analysis should be used to develop work towards the end of Section 3. Anthropogenic controls, spatial differences and relation to CO2 are considered almost independently in Sections 3.5, 3.6 and 3.7. But the spatial differences suggest different processes or source strengths are important in different wind sectors, so these findings should be considered in weekday/weekend differences and the relation to CO2.
Although we don’t have an opportunity to carry out a more detailed analysis of the relationship between FCH4 and the wind direction and the differences between week days and weekends (see our earlier explanations), we decided not to put such summary at the end of section 3. The summary section contains all our main conclusions.

Building heating and traffic are important in winter, but less so in summer when photosynthesis also occurs, and less so for the vegetated sectors. The relation between FCO2 and FCH4 should include a deeper discussion of the underlying processes – and when and where one might expect FCO2 and FCH4 to be correlated or not.
Section 3.7 has been extended. One figure (no. 10) as well as appropriate paragraph has been added:



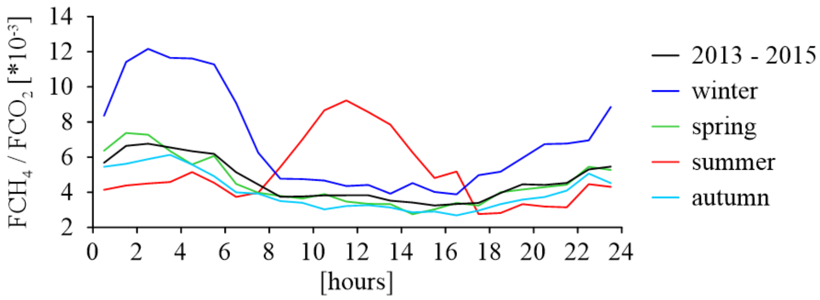


Fig. 10. Monthly FCH4 to FCO2 ratio (up) and mean diurnal courses of FCH$ to FCO2 ratio in the period July 2013 – August 2015 and for seasons.

*"Comparison of FCH4 and FCO2 fluxes allows also the analysis of the relative contribution of each of the fluxes in the total emissions into the atmosphere. The average value of FCH4/FCO2 ratio in 2013-2015 was 3.71*10-3 (Fig. 10, top). Rather stable values of the ratio in months (minimum 2.41*10-3, maximum 5.3*10-3) and the lack of a clear annual course suggest rather comparable magnitude of both fluxes. However, clear diurnal course of the ratio has been observed (Fig. 10, bottom) with reduced values in the day and elevated during night. On average, over the study period and in the transitional seasons, the daily variation of FCH4/FCO2 was rather similar. Between the hours of 9:00 and 17:00, its value was approximately constant of the order 2.5 to 3.5*10-3. At night, these values grow to about 5-7*10-3, which can be explained by a relatively constant methane emissions related to leaks from pipelines, and reduced emission of carbon dioxide which is the result of minimum of traffic load. In winter, the average daily variability of FCH4/FCO2 ratio has been characterized by slightly higher values during the day (about 4.4*10-3) and significantly higher at night reaching 12*10-3 between the hours of 2.00 and 6.00 (Fig. 10, bottom). The cause again can be a minimum of traffic load giving reduced fluxes of FCO2 but also increased methane leaks from pipelines associated with a higher gas consumption for heating of the surrounding buildings. The exception was the daily course FCH4/FCO2 in the summer, which can be described as reverse - the minimum (of the order of 3-5*10-3) was observed at night when maximum (more than 8*10-3) around noon (Fig. 10, bottom). Elevated values of the ratio during the day are the result photosynthesis reducing FCO2 flux."*

The conclusion is strong and provides a good synthesis, setting this work in context.
The comparisons with other sites and other cities is well-written and useful.

P18 L25-7: Can the authors suggest a reason for this?
In our opinion two maximums are reflection of inhabitants diurnal activity - increased consumption of natural gas and traffic rush hours.

Minor comments:

Title: 'results of a 2-year campaign' reads better.
Done

P1 L17-23: In my view this is too much detail for the abstract and 'the measurement

station : : : approximately 1 kilometre' should be omitted.
Done

P1 L30: CH4 and FCH4 are introduced here; to improve readability it may be more helpful to introduce them earlier (or not at all in the abstract), as currently several different ways of referring to the methane flux are used.
Done – FCH4 is now introduced in the end of introduction

P2 L1-2: To improve readability, delete 'The studied area of the centre of Łód´z is also characterised by a cycle of methane exchange – the'
Done

P2 L4: Change 'was characteristic of' to 'occurred in'
Done

P5 L8: Change 'at the same time hinder' to 'do not allow'
Done

P10 L13: Either 'The climate of central Poland is' or 'The climate of Łód´z is' (the reader already knows that Łód´z is in central Poland from P5 L31).
Done

P10 L28: Delete 'Generally, it can be noted that'. (You could say 'generally' before 'prevailed' if it is important.)
Done

P10 L29: Change 'could rarely be' to 'was rarely'
Done

P13 L9: Is 'or' the best word here?
Done

P13 L13: New paragraph after 'hypothetical'
Done

P13 L13: The meaning of this sentence is unclear, suggest deleting.
Done

P14 L4: Suggest deleting 'when they were up to 2.0 g m-2 month-1' as this sounds as though it contradicts the Jan/Feb values. Alternatively change 'up to' to 'around'.
Done

P14 L26: Intended meaning of 'supplementing' is unclear; 'observing' may be more suitable.
Done

P14 L30: Although additional data may be useful for more advanced gap-filling algorithms, simplifications are often used, as is the case in this study. Therefore suggest changing 'are required' to 'may be useful', or similar.
Done

P15 L28: Delete 'mg m-2 day-1'?

Done

P16 L7: Suggest starting this section with 'As mentioned in Section 2.1, building density is: : :'
Done

P16 L23: Missing value.
Done (it shoul be 55-70 nmols/m2s)

P17 L2-7: Delete as this point has been made already in the Introduction.
Done

P17 L9: Change 'should be asked' to 'can be addressed'
Done

P17 L18-20: This sentence is not very precise. Suggest rephrasing to say something like, 'We therefore conclude that FCO2 data cannot be used to facilitate gap-filling of FCH4 data in the centre of Łód´z.'
Done

P17 L32: Change 'in the case' to 'for'
Done

P18 L25: Meaning of 'at the same time' is unclear. Suggest deleting or changing to 'for the same period' if that is the intended meaning.
Done

P18 L25: 'before and after noon' is not very precise and potentially misleading: 'morning and evening' or giving the times would be better.
Done

P19 L1-3: Hinting at the discrepancy between CH4 observations and inventory estimates at this stage leaves the reader suddenly doubting the measurements they have just read about! Therefore this sentence might be more appropriate earlier on (either where the Florence results are discussed or when inventory data are mentioned).
Done (the last sentence has been moved where Florence results are discussed)

P28 Add percentage units to the table or caption.
Done

P31 L2: Delete 'set' Figure captions: 'in the Łód´z centre' can be deleted from most figure captions as it seems unnecessary.
Done

P35 L3: Suggest 'light blue' and 'dark blue' lines.
Done

Technical corrections:

P2 L26: Delete 'the'
Done

P4 L11: Change ‘hang’ to ‘mount’
Done

P4 L13: Change ‘poorly’ to ‘not’ or ‘poorly widespread’ to ‘rare’
Done

P5 L16: Change ‘a turbulent’ to ‘the turbulent’
Done

P6 L1: Delete ‘a’
Done

P12 L25, Pg14 L5: Change ‘twice’ to ‘two times’
Done

P28 L2, P29 L2: Change to ‘July 2013’
Done

P30 L3 Change to ’10.00 to 14.00’
Done

P32 Fig 3: Change y-axis label to ‘air’ (not ‘ait’)
Done

Author reply: Eddy covariance measurements of the net turbulent methane flux in the city centre – results of 2 year campaign in Łódź, Poland by W. Pawlak and K. Fortuniak

Referee 2 comments in bold

We would like to thank referee for his thorough reading of the manuscript and for very detailed, constructive and useful comments, which show his dedication to improving this manuscript.

Pawlak and Foruniak present CH4 flux measurements that were collected over Lodz, Poland for a two year period. Long term measurements of urban CH4 fluxes are still rare; therefore this dataset is of great use to the community. However, I do have some concerns that need to be addressed before this paper is published.

Major comments:

1) The quality of the writing needs to be dramatically improved throughout the paper. I've tried to point out specific areas where improvement is needed; however this list is not exhaustive. I suggest this paper should be thoroughly proofread. Removing repetition and shortening paragraphs will make the paper more readable.

The article has been written in polish and next translated by a professional translator and then sent to a professional native proofreader. <u>Before uploading the revised manuscript we will make every effort to ensure the quality of language is as high as possible</u>. Some repetition has been also removed and shortening paragraphs has been done as well.

2) Source apportionment could be improved, currently it is limited and quite qualitative. Statements such as <u>page 12 line 3</u> and <u>page 13, line 10</u> suggest specific sources, but these are not justified.

As it has been mentioned in the manuscript, we, unfortunately, don't have any inventory data (it is really hard to get such data from the municipal or other offices. Officials think mainly that we try to prove them some irregularities, eg. that leaks from pipelines is their fault and so on. Of course in accordance with the regulations such data should be shared, but they say that we can appeal against their decision). Therefore, our explanation in mentioned above cases are suggestions based on observations.

To improve this I suggest you further examine FCO2 : FCH4 ratios. Currently, these are dismissed due to a lack of strong correlation over daily and monthly averaged timescales. However, strong correlation might not always be expected, CO2 and CH4 have a mixture of common and unique sources. The ratio FCO2 : FCH4 will still give information about the relative abundance of different source types. Whenever a particular source type is suggested you should examine the FCO2 : FCH4 ratio and compare it to literature values for the source.

Section 3.7 has been extended. One figure (no. 10) as well as appropriate paragraph has been added:

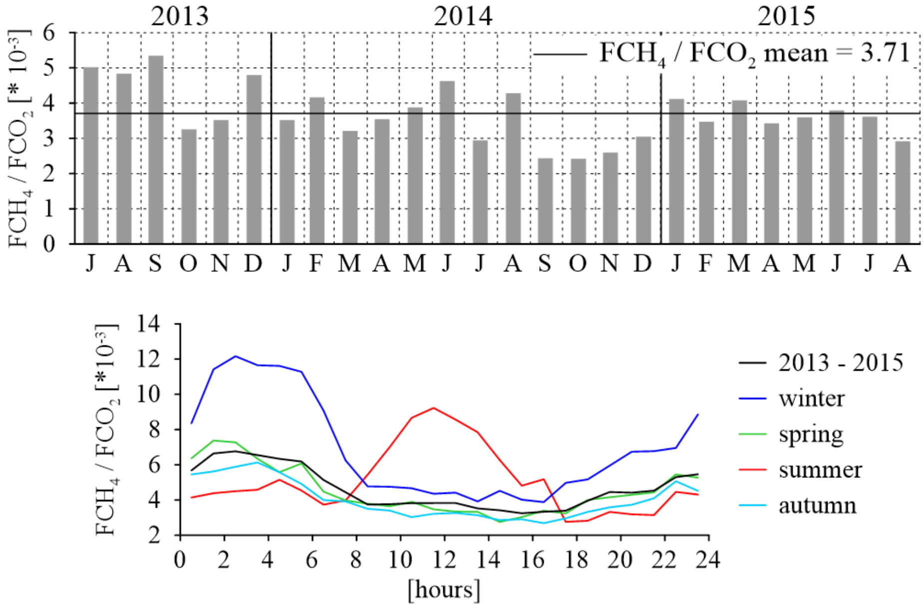


Fig. 10. Monthly FCH4 to FCO2 ratio (up) and mean diurnal courses of FCH$ to FCO2 ratio in the period July 2013 – August 2015 and for seasons.

*"Comparison of FCH4 and FCO2 fluxes allows also the analysis of the relative contribution of each of the fluxes in the total emissions into the atmosphere. The average value of FCH4/FCO2 ratio in 2013-2015 was 3.71*10-3 (Fig. 10, top). Rather stable values of the ratio in months (minimum 2.41*10-3, maximum 5.3*10-3) and the lack of a clear annual course suggest rather comparable magnitude of both fluxes. However, clear diurnal course of the ratio has been observed (Fig. 10, bottom) with reduced values in the day and elevated during night. On average, over the study period and in the transitional seasons, the daily variation of FCH4/FCO2 was rather similar. Between the hours of 9:00 and 17:00, its value was approximately constant of the order 2.5 to 3.5*10-3. At night, these values grow to about 5-7*10-3, which can be explained by a relatively constant methane emissions related to leaks from pipelines, and reduced emission of carbon dioxide which is the result of minimum of traffic load. In winter, the average daily variability of FCH4/FCO2 ratio has been characterized by slightly higher values during the day (about 4.4*10-3) and significantly higher at night reaching 12*10-3 between the hours of 2.00 and 6.00 (Fig. 10, bottom). The cause again can be a minimum of traffic load giving reduced fluxes of FCO2 but also increased methane leaks from pipelines associated with a higher gas consumption for heating of the surrounding buildings. The exception was the daily course FCH4/FCO2 in the summer, which can be described as reverse - the minimum (of the order of 3-5*10-3) was observed at night when maximum (more than 8*10-3) around noon (Fig. 10, bottom). Elevated values of the ratio during the day are the result photosynthesis reducing FCO2 flux."*

Specific comments:

Abstract

Page 1, line 11 to 12- Reword e.g. "Long-term continuous measurements of CH4 fluxes from cities are still relatively rare."

Done

Page 1, lines 11- replace "turbulent exchange" with "flux" here and throughout.

Done

Page 1, lines 17 to 20 contain two much detail for an abstract. You don't need to give manufacturers and part numbers. Saying you used the eddy covariance technique should be sufficient.

Information about instrumentation and manufactures has been removed from this section

Page 1, line 24 to 27- These two sentences should be simplified and merged. It is sufficient to say that the centre of Lodz is found to be a net source of methane to the atmosphere.
Two sentences has been merged into one: "The results show that positive methane fluxes definitely dominate which indicates that the study area of the centre of Łódź is a net source of methane to the troposphere."

Page 1, line 26- give summer mean flux.
Done, now this part of abstract reads:
"The measurements also indicated the existence of a clear annual rhythm of the turbulent flux of methane in the centre of Łódź. On average, the values observed in winter amounted to ~40-60 nmol·m-2·s-1 and were significantly larger than in summer (~20 nmol·m-2·s-1)."

Introduction
The introduction is too long and incoherent. This section could do with a complete re-write, focusing on
1) what sources of methane are important in urban areas
2) what are the challenges with determining urban emissions (e.g. spatiotemporal variability, lack of long term measurements)
3) what techniques can be used to determine fluxes (EC, inverse models, etc)
4) how do your measurements help address these issues.
The introduction has been rewritten, shortened and clarified regarding to referee's suggestions mentioned above. We would like to leave information about methane sources outside the city to emphasize a significant difference between processes and source of methane in urban and rural areas.

Introduction:
"The temporal and spatial variability of greenhouse gas fluxes in the atmosphere is at present one of the most widely discussed climatological problems. Methane, despite its trace presence in the air (ca 1.8 ppm, Hartman et al., 2013), plays an important role in the environment. It participates in the global carbon cycle and is considered one of the greenhouse gases whose concentration in the atmosphere affects the radiation balance of the Earth's surface. An increase in the concentration of methane contributes to an enhancement of the greenhouse effect; therefore, the emissions of this gas to the atmosphere should be carefully monitored.
Methane is produced during the process of methanogenesis under anaerobic conditions, from the decay of organic plant debris in water. The most important source of methane in the world is wetlands (Shurpali et al., 1998; Rinne et al., 2007; Baldocchi et al., 2012; Hatalaa eta al., 2012), but also paddy fields (Miyata et al., 2000), cattle farming (Laubach and Kelliher, 2005; Dengel et al., 2011; Hartmann et al., 2013; Nicollini et al., 2013), as well as emissions from the soil (Smeets et al., 2009; Denmead et al., 2010; Wang et al., 2013). Moreover, emissions of methane accompany forest fires and grass vegetation. The effect of the combustion of natural gas (which contains at least 80% methane) is mainly water vapour and carbon dioxide. The combustion of fossil fuels is, however, predominantly incomplete, and therefore it is an important factor causing anthropogenic methane emissions. This happens in the case of combustion of both natural gas and hydrocarbons contained in petrol and other fuels (Nam, 2004; Nakagawa et al., 2005; Wennberg et al., 2012). Another important source of methane in urbanized areas is leakage from urban gas pipelines (Lowry, et al., 2001; Gioli et al., 2012; Wennberg et al., 2012; Phillips et al., 2013). Methane may also be emitted during the anaerobic respiration of bacteria in urban soils (Bogner and Matthews, 2003) and in the course of decomposition of solid waste and wastewater in sewage systems and at landfill sites (Bogner and Matthews, 2003; Laurila et al., 2005; Lohila etal., 2007; Wennberg et al., 2012; Jha etal., 2014). On the other hand, certain soil bacteria consume methane, which is one of the processes of its removal

from the air (Goldman et al., 1995; Kaye et al., 2004; Groffman et al., 2006; Groffman and Pouyat, 2009). Methane is involved in some of the reactions leading to photochemical smog formation (Seinfeld and Pandis, 2006). The disintegration of methane also results from its reacting with the hydroxyl group in the atmosphere (Whalen, 2005). Annual global emission of methane to the atmosphere has been estimated as ~5000 Tg of CH4, and emission from landfills and waste (87-94 TG of CH4) or fossil fuels (85-105 Tg of CH4) are 2-3 times lower than estimated emission form wetlands (177-284 Tg of CH4) (Ciais et al, 2013).

Research on the methane content in the air is now a priority because, as it follows from the literature on the problem, the city may be a significant source of this gas (Elliot et al., 2000; Gioli et al. 2012; O'Shea et al., 2012; Nicolini et al., 2013; Phillips et al., 2013; Christen, 2014; Kumar and Sharma, 2014). The measurements of changes in CH4 concentrations have been carried out for decades (Ciais et al., 2013; Hartmann et al., 2013), while the analyses of its flux, especially in urban areas, are extremely rare. In recent years, there have been approximately 500 stations measuring the fluxes of CO2 around the world, of which only ca 20 are located in cities and only a few were able to measure methane flux (Nordbo et al., 2012; Oliphant, 2012; Christen, 2014). It can be concluded that the measurements of methane fluxes in the cities are in the early stage and there are still some challenges like long term measurements (much longer than a few weeks or months) and relationship between methane fluxes and land use.

Basics of theory and measurement techniques of turbulent exchange of mass, energy and momentum fluxes have been developed for decades (Stull, 1988; Lee et al., 2005; Foken, 2008; Aubinet et al., 2013). The measurements of the fluxes of methane were severely limited due to the lack of suitable sensors which to have appeared a few years ago (Pattey et al. 2006; Hendricks et al., 2008; Eugster and Pluss, 2010; Dengel et al., 2011; Detto et al., 2011; Sakabe et al., 2012). At present, the most widely used instrument is in all probability the LI7700 Open Path CH4 Analyzer (Burba and Anderson, 2010; McDermitt et al., 2011) and eddy-covariance as a measurement technique (Aubinet et al., 2012). All over the globe, there are only a few long-term, continuous measurement series of turbulent fluxes of water vapour and carbon dioxide recorded in urban areas (Christen, 2014). In the case of methane flux, such series are probably at the implementation phase, since previous studies focused on areas which are the largest source of methane, i.e. natural wetlands (Shurpali et al., 1998; Rinne et al., 2007; Baldocchi eta al., 2012; Hatalaa eta al., 2012; Aubinet et al., 2013), agricultural land (paddy fields, Miyata et al., 2000) or over forests (Smeets et al., 2009; Wang et al., 2013). The chamber method, widely used in rural areas, has only a limited relevance in the city: it makes it possible to take measurements of methane emissions from the specific areas like urban lawns (Baciu et al., 2008), however, it cannot be used in the case of larger urban areas. A variety of techniques have recently been applied to provide independent estimates of urban CH4 emissions like airborne observations (O'Shea et al., 2014; Mays et al., 2009), Fourier Transform Spectrometry (Wunch et al., 2009) or isotopic source apportionment studies (Lowry et al., 2001). Morizumi (1996), in turn, suggested the occurrence of covariability of radon Rn-222 and the methane flux concentrations, which, based on this, he estimated to be 20 mg·m-2·day-1. In Poland, the issue of exchange of greenhouse gases in an urban area is studied, besides Łódź, in Cracow where, based on the measurements of CH4 concentrations and the height of the atmospheric boundary layer, the average monthly nocturnal flux of methane has been estimated to be 0.8 do 3 mg·m-2·h-1 (Kuc et al., 2003; Zimnoch et al., 2010).

The aim of this study is to analyze the temporal variability of the turbulent flux of methane (FCH4) based on a long-term series of measurements recorded for over two years in the centre of Łódź between July 2013 and August 2015. The diurnal variability of FCH4 was analysed and monthly values of the flux were determined and an attempt was undertaken to assess the cumulative annual exchange of methane between an urban area and the troposphere in order to determine whether the centre of Łódź was an equally efficient source of methane to the troposphere as of carbon dioxide. The measurement results were compared to the variability of selected meteorological elements. As the methane emissions in the city are determined mainly by anthropogenic factors, the values of *fluxes on weekdays and at weekends were compared. Due to the impossibility to obtain relevant data, there was no comparison made with the values of fluxes using specific inventory methods. "*

Page 2, line 11- "The temporal and spatial exchangeability of the concentration of greenhouse gases" is confusing- simplify e.g. "The temporal and spatial variability of greenhouse gas fluxes: : :".
Done

Page 2, lines 12-20- This section should be made more concise, much of the information given is very general.
This part of manuscript has been rewritten: "Methane, despite its trace presence in the air (ca 1.8 ppm, Hartman et al., 2013), plays an important role in the environment. It participates in the global carbon cycle and is considered one of the greenhouse gases whose concentration in the atmosphere affects the radiation balance of the Earth's surface. An increase in the concentration of methane contributes to an enhancement of the greenhouse effect; therefore, the emissions of this gas to the atmosphere should be carefully monitored."

Page 2, line 20 to Page 3 line 12- I would only focus on the sources important in urban areas and then give an estimate of their proportion of total global emissions.
We would like to post information about the different(not only urban) sources of methane to the atmosphere, to emphasize a significant difference between processes and source of methane in urban and rural areas. Information: „ Annual global emission of methane to the atmosphere has been estimated as ~5000 Tg of CH4, and emission from landfills and waste (87-94 TG of CH4) or fossil fuels (85-105 Tg of CH4) are 2-3 times lower than estimated emission form wetlands (177-284 Tg of CH4) (Ciais et al, 2013)." has been added to the manuscript.

Page 3 line 13-15- This sentence is very confusing, please reword.
This sentence has been reworded: "Research on the methane content in the air is now a priority because, as it follows from the literature on the problem, the city may be a significant source of this gas (Elliot et al., 2000; Gioli et al. 2012; O'Shea et al., 2012; Nicolini et al., 2013; Phillips et al., 2013; Christen, 2014; Kumar and Sharma, 2014)."

Page 3 line 17-18- Reword "The classical measurements: : :"
This sentence has been reworded: "The measurements of changes in CH4 concentrations have been carried out for decades (Ciais et al., 2013; Hartmann et al., 2013), while the analyses of its flux, especially in urban areas, are extremely rare."

Page 3 line 20-25- Reword/simplify this sentence is currently very confusing.
This part of manuscript has been simplified: "Basics of theory and measurement techniques of turbulent exchange of mass, energy and momentum fluxes have been developed for decades (Stull, 1988; Lee et al., 2005; Foken, 2008; Aubinet et al., 2013)."

Page 3 line 25 Change "suitable instruments" to "precise fast response instruments"
Done

Page 4 line 13- "poorly widespread" please reword.
Done, this sentence has been rewritten: "The complicated methodology resulting from the heterogeneity of urban areas and the necessity to mount the sensors at least several tens of meters above the ground, as well as considerable funds necessary to launch a measurement station caused the fact that the measurements of turbulent fluxes are still not widespread."

Measurement site and instrumentation

This section needs to include an assessment of the measurement uncertainty. You need to give precisions for the CH4 and meteorological variables.
Precision of CH4 and u, v, w measurements has been added to the text. Since all the meteorological sensors were produced by well-known manufacturers (Vaisala, Campbell Scientific, Vector), we are not sure that the giving detailed information about their accuracy is really required.

Page 6, line 4 to 5- "which definitely facilitates investigating the climate of the city". This is vague. Are you trying to say that there are suitable conditions to apply the eddy covariance technique.
No, in this sentence we just try to prove that Łódź's surroundings not have "negative" influence on the local climate e.g. sea breeze, local mountains winds, etc.

Page 7, line 20-21-"using a standard measurement kit". This is vague. Reword/Remove.
This sentence has been reworded: "The measurements of the turbulent fluxes of methane were carried out using a standard measurement set consisting of an ultrasonic anemometer RMYoung model 81000 (RMYoung, Traverse City, Michigan, USA,) and a methane fluctuation sensor with an open measurement path LI7700 (Li-cor, Lincoln, Nebraska, USA)."

Page 8, line 2- "slightly lower"- Give the distance.
It is 30 cm, information about it has been added

Page 8, line 8- The word "fluctuations" is vague, change to "mole fractions"/ "concentrations" whichever is appropriate. There are several other occasions where this is used and should be changed.
Done

Page 8, line 9- How much did the zero and span change? Did you do any calibrations?
There was no and zero and span changes

Page 9, line 2- This sentence is not necessary.
The sentence has been removed

Page 9, line 12- Are you able to show a power spectrum/cospectrum to support this?
We've measured turbulent fluxes over urbanized area since year 2000. At the beginning we focused at the turbulent components of energy balance (sensible and latent heat flux), next CO2 flux and recently CH4 flux. That time we have checked optimal averaging period. The similar to Van der Hoven (1957) wind speed spectrum (Fig.1 – both figures form Fortuniak (2010)) shows minimum for frequencies around 1h^{-1}. We have also compared the turbulent fluxes calculated for 1 h averaging period with mean of 4 values for 15 min averaging period (Fig. 2). Results show that 15 min averaging period can underestimate turbulent fluxes about 4-6% (4% in the case of QH and 6% for QE). Moreover, the increase of measurement height shifts spectrum toward lower frequencies (Fokken et al. 2012, p. 15). Thus, the relatively high elevation of the sensors in our case (37 m) suggests the need of the extension of the averaging period to avoid low-frequency spectral losses.

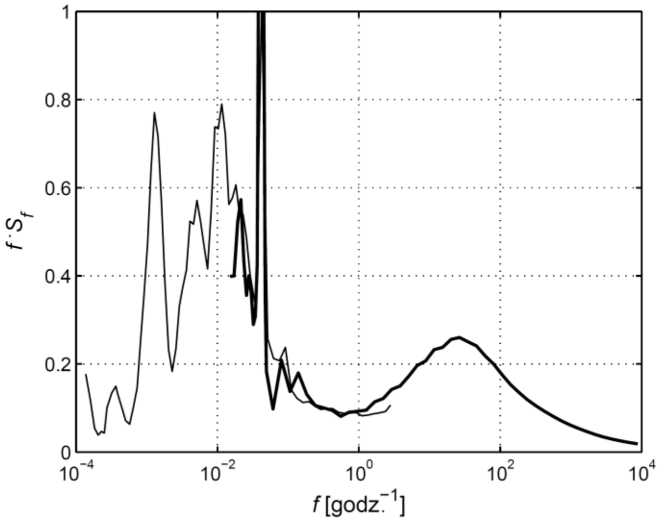


Fig. Wind speed spectrum at Lipowa measurement station in Łódź. Left thin line – on the base of cup anemometer (10 min data from years 2001 -2002), Right bolt line – on the base of sonic anemometer data (10 Hz) from the period 12.06.2002–4.08.2002. ("godz." means "hour" in Polish)

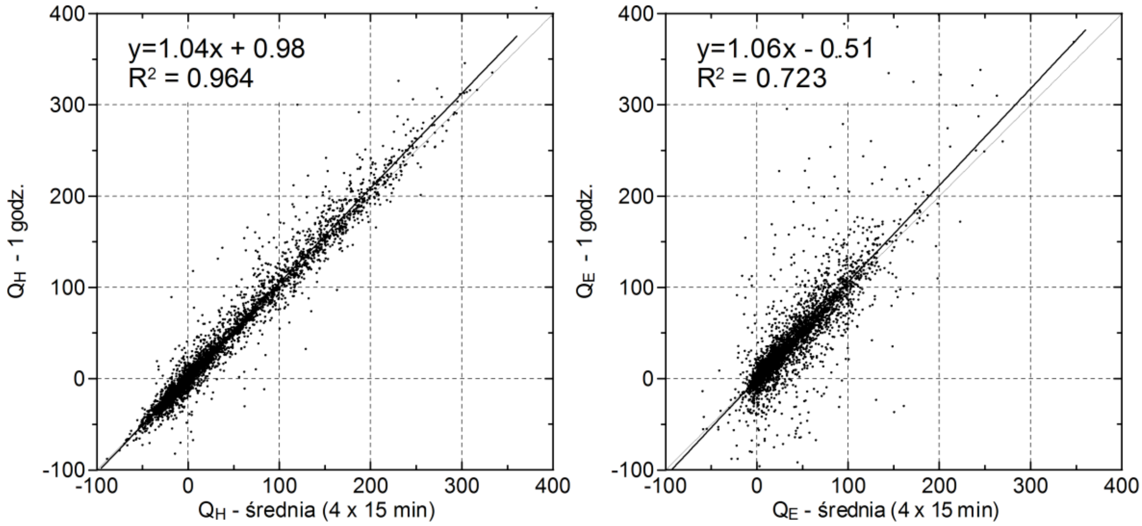


Fig. 2 The comparison of turbulent fluxes of sensible, QH, and latent, QE heat calculated for 1 hour averaging period (Q – 1 godz.) and as a mean of 4 values for 15-min averaging period (Q – średnia (4x15min)). Lipowa measurement station years 2000-2002.

Fortuniak, K., 2010, *Radiacyjne i turbulencyjne składniki bilansu cieplnego terenów zurbanizowanych na przykładzie Łodzi*, Wyd. UŁ, Łódź, 232 pp (in Polish) http://nargeo.geo.uni.lodz.pl/~meteo/kf/publikacje_kf_PDF/r2010_KFortuniak_Radiacyjne_i_turbulencyjne_all.pdf .

Foken T, Aubinet M, Leuning (2012) The eddy covariance method, in: Aubinet M, Vesala T, Papale D (Eds.), *Eddy Covariance: A Practical Guide to Measurement and Data Analysis, Partitioning of net fluxes*, Springer, Dordrecht, Heidelberg, London, New York. pp. 1–19.

Page 9 – please use shorter paragraphs.

According to referee comments this section has been divided to smaller paragraphs

Results

Page 11, line 23- "was a definite domination"- please reword.

Done: "First of all, regardless of the season, mainly positive of FCH4 were observed (Fig. 3)."

Page 11, line 24-

What is the reason for the negative fluxes for 6-7% of the time? Is it a measurement artefact or a sink in the flux footprint?

Unfortunately we can't answer to this question. In our opinion it is not a sink influence but we also cannot prove that the negative values occur because of some artefact or "strange" results of calculations. These negative values occur during whole year independently from season.

Page 14, line 2-3- This is confusing please clarify.

Done: "Based on the average daily patterns of FCH4 calculated for each month (the sum of the average hourly FCH4 multiplied by the number of days in the month), the exchange of methane in the successive months of the study period was determined (Fig. 5)."

Page 14, line 6- "twice lower". Reword e.g. "During the summer CH4 fluxes decreased by greater than a factor 2 to : : :".

This sentence has been rewritten: "The summer values were more than two times lower and dropped to 0.7-0.8 g·m-2·month-1."

Page 14, line 20-22- This is not necessarily correct, the seasonal cycle of background CH4 is largely due to changes in OH.

Information about it has been added to the text: "The high winter values of the flux of methane are accompanied by higher concentrations of the gas in the air and seasonal changes in OH concentration."

Page 14, line 32- I am not sure how you did this could you be more precise?

Altgough there is no precise methods for FCH4 data gapfilling in the urban areas simple method has been used:

1) *Annual FCH4 flux has been calculated on the basis of mean diurnal courses of FCH4 in months. (mean FCH4 at 0 am x number of days in selected months + mean FCH4 at 1 am x number of days in selected months +…. mean FCH4 at 11 pm x number of days in selected months and the next the accumulation of monthly totals)*
2) *Simple gap filling – if the gap was short (not longer than 3 hours) interpolation has been used. If the gap was longer it was filled with the data taken from mean hourly value of FCH4 in respective month.*

These procedure has been explained in the text

Page 17, line 28-30- In the previous section you suggested there was a weak relationship between CO2 and CH4.

Yes, because there is rather weak relationship between CO2 and CH4 but both have an annual cycle.

Summary

Page 18, line 4-5- I don't think this is necessary to say.

This sentence has been removed

Page 18 line 21- The flux given by O'shea et al. is just for 1 day for comparison with their aircraft measurements.

Yes, information about it has been removed and results by Helfter et al. (2016) has been added

Page 18 line 14- You should mention studies on urban CH4 emissions using techniques other than eddy covariance, e.g. Peischl, J., et al. (2013), Quantifying sources of methane using light alkanes in the Los Angeles basin, California, J. Geophys. Res. Atmos., 118, 4974–4990, doi:10.1002/jgrd.50413. Mays, K. L., Shepson, P. B., Stirm, B. H., Karion, A., Sweeney, C., and Gurney, K. R.: Aircraft based measurements of the carbon footprint of Indianapolis, Environmental Science and Technology, 43, 7816-7823, doi:10.1021/es901326b, 2009 Wunch, D., P. O. Wennberg, G. C. Toon, G. Keppel-Aleks, and Y. G. Yavin (2009), Emissions of greenhouse gases from a North American megacity, Geophys. Res. Lett., 36, L15810, doi:10.1029/2009GL039825
Suggested above information has been added as well as results from the latest paper by Helfter et al (ACPD under review): "Based on the existing measurements, it is difficult to attempt to seek a similar dependence for the flux of methane. Since now only in London relationship between FCH4 and population has been found (Helfter et al., 2016). There are also several published results of urban CH4 emissions obtained with other than eddy-covariance technique like with usage of alkanes (Los Angeles, Pieschl et al., 2013), aircraft measurements (Indianapolis, Mays et al., 2009) or ground-based Fourier transform spectrometer (Los Angeles, Wunch et al., 2009). All of them reports existence of higher FCH4 fluxes than measured in Łódź."

Page 19 line 1-3- This is very vague, either expand or remove.
Done

Table 1 – Based on page 10, line 9, I assume these are percentages. Please clarify?
Yes these are percentages, the table has been corrected

Figure 9. For the daily mean plot I would've expected over 700 data points for a two year period. There appears to be much less, have I missed something?
Yes, the number of points is much less. Although there is no proper FCH4 gap filling procedure for urban sites the daily means has been calculated only for days with 75% of good data.

Eddy covariance measurements of the net turbulent methane flux in the city centre – results of 2 ~~years~~year campaign in Łódź, Poland.

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Abstract

In the period ~~between~~ July 2013 ~~and~~to August 2015, continuous measurements of turbulent methane exchange between an urbanised area and the atmosphere were carried out in Łódź. ~~Such long,~~Long-term continuous ~~measurement series~~measurements of ~~turbulent methane exchange between the city and the atmosphere~~CH_4 fluxes from cities are still ~~a rarity.~~relatively rare. The measurement station was located in the centre of the city, where since 2000 fluxes of energy (sensible and latent heat) and since 2007 fluxes of mass (carbon dioxide) have been continuously measured~~ since 2000 and 2007, respectively~~. In the immediate vicinity of the measurement station there are potential sources of methane, such as ~~streets with~~ vehicle traffic~~ or~~, dense sewerage infrastructure and natural gas networks. To ~~determine the~~assess fluxes, the eddy covariance technique was used~~; the measurement station was equipped with instruments for recording fluctuations in the vertical component of the wind speed (an ultrasonic 3D anemometer, RM Young 81000, RM Young, USA) as well as the concentration of methane in the air (an open path Li 7700 CH4 Analyzer, Li-cor, USA).~~. The ~~devices were mounted on a mast at a height of 37 metres above ground level and, on average, 20 metres over the roofs of the surrounding buildings. The results were therefore averaged for an area with a diameter of approximately 1 kilometre. Our~~ study aim was to investigate the temporal variability of ~~the turbulent exchange of~~ methane fluxes in the city-atmosphere system.

The results show ~~in the first place~~ that positive methane fluxes ~~(turbulent gas transport from the surface to the atmosphere) definitely~~ dominate ~~compared with negative fluxes. This~~which indicates that the ~~study area of the~~ centre of Łódź is a net source of methane to the

troposphere. The measurements also ~~indicated~~indicate the existence of ~~a clear~~an annual rhythm ~~of~~in the turbulent flux of methane~~ in the centre of Łódź (on~~. On average, the values observed in winter amounted to ~40-60 nmol·m-2·s-1 and were significantly larger than in summer~~).~~ (~20 nmol·m^{-2}·s^{-1}). The daily variability ~~of~~in the methane flux ~~of CH$_4$ (FCH$_4$)~~ is ~~faintly~~also just visible throughout the year. ~~The studied area of the centre of Łódź is also characterised by a cycle of methane exchange – the values~~Values measured on working days were higher ~~by~~ (6.6% (winter) to 5.6% (summer~~)~~)) than those observed at weekends. The largest monthly exchange ~~was characteristic of~~occurred in the winter months (from 2.0 to 2.7 g·m^{-2}·month^{-1}) and the lowest occurred in summer (from 0.8 to 1.0 g·m^{-2}·month^{-1}).

The mean daily patterns of ~~FCH$_4$~~methane fluxes in consecutive months were used to determine the cumulative annual exchange. In 2014, the centre of Łódź emitted a net quantity of almost 18 g·m^{-2}. Furthermore, the study ~~analyses~~assessed the ~~covariability~~co-variability of methane and carbon dioxide fluxes.

1 Introduction

The temporal and spatial ~~exchangeability of the concentration~~variability of greenhouse ~~gases~~gas fluxes in the atmosphere is ~~at present~~currently one of the most widely discussed climatological problems in the scientific community. Methane, despite its trace presence in the air (ca 1.8 ppm, Hartman et al., 2013), plays an important role in the ~~energy exchange between the biosphere, the atmosphere, the lithosphere and the hydrosphere. In the first place, it~~environment. It participates in the global carbon cycle~~; furthermore, besides water vapour, carbon dioxide, nitrous oxide, and CFCs, it is considered~~ and is one of the greenhouse gases whose concentration in the atmosphere affects the ~~formation of the~~ radiation balance of the ~~Earth's~~earth's surface. An increase in the concentration of methane contributes to an enhancement of the greenhouse effect~~; therefore, the~~ (Ciais et al., 2013). Therefore, emissions of this gas to ~~the~~ atmosphere should be carefully monitored.

Methane is produced during the process of methanogenesis under anaerobic conditions, from the decay of organic plant debris in water. The most important source of methane in the world is wetlands (Shurpali et al., 1998; Rinne et al., 2007; Baldocchi et al., 2012; Hatalaa eta al., 2012), but paddy fields (Miyata et al., 2000), cattle ~~farming~~farms (Laubach and Kelliher, 2005; Dengel et al., 2011; Hartmann et al., 2013; Nicollini et al., 2013~~),~~) and emissions from the soil are all important sources (Smeets et al., 2009; Denmead et al., 2010; Wang et al.,

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2013). Moreover, ~~the~~ emissions of methane accompany forest fires and grass vegetation~~, and methane is also the main component of natural gas~~. The effect of the combustion of natural gas (which contains at least 80% methane) is mainly water vapour and carbon dioxide. The combustion of fossil fuels is, however, predominantly incomplete, and is therefore an important factor causing anthropogenic methane emissions. This happens in the case of combustion of both natural gas and hydrocarbons contained in petrol and other fuels (Nam, 2004; Nakagawa et al., 2005; Wennberg et al., 2012). Another important source of methane in urbanised areas is leakage from urban gas pipelines (Lowry, et al., 2001; Gioli et al., 2012; Wennberg et al., 2012; Phillips et al., 2013). Methane may also be emitted during the anaerobic respiration of bacteria in urban soils (Bogner and Matthews, 2003) and in the ~~course of~~ decomposition of solid waste and wastewater in sewage systems and at landfill sites (Bogner and Matthews, 2003; Laurila et al., 2005; Lohila etal., 2007; Wennberg et al., 2012; Jha etal., 2014). ~~On the other hand, certain soil bacteria consume~~In contrast, methane~~, which~~ is ~~one of the processes of its removal~~removed from the air by consumption by soil bacteria (Goldman et al., 1995; Kaye et al., 2004; Groffman et al., 2006; Groffman and Pouyat, 2009). ~~Furthermore, methane~~Methane is involved in some of the reactions leading to photochemical smog formation (Seinfeld and Pandis, 2006). The disintegration of methane also results from its reacting with the hydroxyl group in the atmosphere (Whalen, 2005). Annual global emissions of methane to the atmosphere have been estimated as ~5000 Tg. Emissions from landfills and waste (87-94 TG) or fossil fuels (85-105 Tg) are 2-3 times lower than estimated emission from wetlands (177-284 Tg) (Ciais et al, 2013).

Research into the methane content in the air is now a priority~~, not only from the point of view of the natural sciences, but also that of the economic and social sciences (Hartmann et al., 2013), and, as it follows from~~ because the literature ~~on the problem, the city may~~indicates that cities could be a significant source of this gas (Elliot et al., 2000; Gioli et al. 2012; O'Shea et al., 2012; Nicolini et al., 2013; Phillips et al., 2013; Christen, 2014; Kumar and Sharma, 2014). The ~~classical~~ measurements of changes in ~~CH_4 concentrations~~ methane concentration have been carried out for decades (Ciais et al., ~~2013; Hartmann et al., 2013), while~~ 2013; Hartmann et al., 2013). However, the analysis of its flux in urban areas is extremely rare. In recent years, there have been approximately 500 stations measuring the fluxes of carbon dioxide (CO_2) around the world of which approximately 20 are located in cities and only a few are able to measure methane flux (Nordbo et al., 2012; Oliphant, 2012; Christen, 2014; Helfter et al., 2016). It can be concluded that the measurement of methane flux in cities is in its infancy and challenges like the need for long term measurements (beyond a month) and the ~~analyses of its exchange, especially in urban areas, are extremely rare.~~ relationship between methane fluxes and land use are yet to be overcome.

The ~~process~~development of the ~~exchange of methane and other greenhouse gases between the ground and the atmosphere is closely related to the turbulent air movement in the atmospheric boundary layer,~~theory and ~~therefore such~~ measurement techniques ~~should be used in the research which allow the determination~~of turbulent exchange of ~~vertical turbulent~~ mass, energy and momentum fluxes~~, and which~~ have been ~~developed~~progressing for decades (Stull, 1988; Lee et al., 2005; Foken, 2008; Aubinet et al., ~~2013). Unfortunately, the lack of suitable instruments resulted in the fact that it was initially only possible to use indirect methods employing empirical coefficients selected arbitrarily by the researchers, or not allowing for the turbulent mixing of air in the boundary layer, such as the gradient method or the chamber method (Nicolini et al., 2013). The instruments to measure the turbulent fluxes of greenhouse gases such as water vapour and carbon dioxide became more widely available several years ago and since then (the first half of the 1990s) the research has intensified (Aubinet et al. 2012). Unfortunately, the measurements of the turbulent exchange of methane were~~ 2013). Historical measurements of methane flux have been severely limited due to the lack of suitable sensors~~,~~ which ~~did not begin to appear until a few years ago~~have only recently become available (Pattey et al. 2006; Hendricks et al., 2008; Eugster and Pluss, 2010; Dengel et al., 2011; Detto et al., 2011; Sakabe et al., 2012). At present, one of the most widely used ~~instrument~~instruments is ~~believed to be~~ the ~~LI-7700~~LI7700 Open Path CH_4 ~~Analyzer~~Analyser (Burba and Anderson, 2010; McDermitt et al., 2011~~). The number of publications describing the results of measurements of the turbulent flux of the gas is therefore relatively small, in contrast to the turbulent process of carbon dioxide exchange which has been described fairly specifically for more than a decade; however, it should be noted that this kind of research in urbanised areas is still relatively rare (Nordbo et al., 2012; Oliphant,~~) which uses eddy-covariance as a measurement technique (Aubinet et al., 2012). ~~In recent years, there have been approximately 500 stations measuring the turbulent exchange of CO_2 around the world, of which only ca 20 are located in cities (Nordbo et al., 2012; Oliphant, 2012; Christen, 2014). The complicated methodology resulting from the heterogeneity of urban areas and the necessity to hang the sensors at least several tens of metres above the ground, as well as considerable funds necessary to launch a measurement station have meant that measurements of turbulent fluxes are still poorly widespread.~~ Worldwide, there are only a few long-term, continuous measurement ~~series of~~stations measuring turbulent fluxes of water vapour and carbon dioxide ~~recorded~~ in urban areas (Christen, 2014~~). In the case of~~; Helfter et al., 2016). For methane flux in urban areas, such ~~series~~data are probably at the implementation phase~~,~~ ~~since~~ because previous studies focused on areas which are the largest source of methane~~,~~ i.e. natural wetlands (Shurpali et al., 1998; Rinne et al., 2007; Baldocchi eta al., 2012; Hatalaa eta al., 2012; Aubinet et al., 2013), agricultural land (paddy fields, Miyata et al., 2000) or forests (Smeets et al., 2009; Wang et al., 2013). The chamber method, widely used in rural areas, has only ~~has~~ a limited relevance in the city~~: it~~. This method makes it possible to take ~~punctual~~ measurements of methane emissions from specific areas like urban lawns (Baciu et al., 2008~~);~~), however, it cannot be used in ~~the case of~~ larger urban areas. ~~The few published results of measurements of methane fluxes in urbanised areas indicate their positive values, i.e. the emissions of methane into the atmosphere. In Germany, based on various kinds of indirect methods, the existence of a flux of methane into the atmosphere was demonstrated, comparable to those observed in wetlands. The leakage of gas from the natural gas system is suggested as the main reason for the existence of the flux (Shorter et al., 1996).~~A variety of techniques have recently been applied to provide independent estimates of urban methane emissions such as airborne observations (O'Shea et al., 2014; Mays et al., 2009), Fourier Transform Spectrometry (Wunch et al., 2009) or isotopic source apportionment studies (Lowry et al., 2001). Morizumi (1996~~), in turn,~~) suggested the occurrence of covariability of radon Rn-222 ~~radon~~ and ~~the~~ methane flux concentrations~~,~~ which~~, based on this, were~~ he estimated to be 20 mg·m^{-2}·~~24h^{-1}. In recent years, there have been studies showing the results of measurements performed in Florence (Gioli et al. 2012) and London (O'Shea et al., 2012). The measurements taken in Columbus, USA, enabled the estimation of methane exchange between wetlands located in the city and the troposphere (Morin et al., 2015).~~day^{-1}. In Poland, the issue of exchange of greenhouse gases in an urban area is studied~~, besides Łódź,~~ has also been studies in Cracow~~,~~ where, based on the measurements of ~~CH_4~~methane concentrations and the height of the atmospheric boundary layer, the average monthly nocturnal flux of methane has been estimated to be 0.8 ~~to~~do 3 mg·m^{-2}·h^{-1} (Kuc et al., 2003; Zimnoch et al., 2010). ~~The question of determining the features of diurnal and seasonal variability of the vertical turbulent exchange of methane in the city and the assessment of the impact of meteorological conditions on the exchange intensity of the gas should be regarded as still open. The measurements described so far, despite having contributed valuable information, at the same time hinder an estimate of the annual flux of methane emitted by urban areas.~~

~~The measurements of turbulent fluxes of mass and energy have been taken in the centre of Łódź since the autumn of 2000 (Offerle et al., 2006a; 2006b, Fortuniak et al., 2013, Fortuniak and Pawlak, 2014). At present, the continuous series of measurements of greenhouse gas fluxes cover 15 years (water vapour, with an interval in the years 2004-2005), and 9 years in the case of carbon dioxide (Pawlak et al., 2011). In July 2013, the measurement kit was equipped with a methane gas analyzer (Li 7700 open-path CH4 Analyzer, Li-cor, USA).~~

The aim of this study is to analyse the temporal variability of ~~a~~the turbulent flux of methane (FCH$_4$) based on a long-term series of measurements recorded for over two years in the centre of Łódź ~~between~~ (July 2013 ~~and~~to August 2015~~. Furthermore, the~~). The diurnal variability of FCH$_4$ was analysed ~~in the following months, the~~and monthly values of the flux were determined~~ and an attempt was undertaken to assess~~. An assessment of the cumulative annual exchange of methane between ~~an~~ urban ~~area~~Łódź and the troposphere ~~in order~~was completed to determine whether ~~the centre of Łódź~~it was an equally efficient source of methane to the troposphere as of carbon dioxide. The measurement results were compared to the variability of selected meteorological elements. As the methane emissions in the city are determined mainly by anthropogenic factors, the ~~values~~value of fluxes on weekdays and at weekends were compared. ~~Due to the impossibility to obtain relevant data, there was no~~No comparison was made ~~with~~to the ~~values of~~ fluxes estimated using specific inventory methods~~.~~ because of a lack of data.

2 Measurement site and instrumentation

2.1 Study area and site location

Łódź is one of the largest cities in Poland. The area of the city is about 295 square kilometres (km^2~~,~~), and its population is estimated at ~~about~~ 706~~ thousand~~,000 residents. The city is located in central Poland~~,~~ on ~~a~~ relatively flat terrain ~~sloping~~which slopes south-westwards~~ (its~~. Its altitude ~~varying~~varies from ~~∼~~280 to ~~∼~~160 m~~.a.s.l.).~~ above mean sea level. The most densely built-up city centre area covers ~~an area of~~ 80 km^2 and the altitude ~~differences~~difference in this part of ~~town do~~the city does not exceed 60 m. In the immediate vicinity of Łódź, there are no large bodies of water, rivers or orographic obstacles~~, which definitely facilitates investigating~~ impacting on the climate of the city which are worthy of investigation. Another factor making it easier to take measurements of turbulent fluxes of mass and energy in Łódź, is that the city,

~~unlike other large cities in Poland,~~ does not have a standard central sector of tall buildings, ~~clearly~~ towering over ~~the~~an urban canopy layer~~.~~ unlike other large cities in Poland.

The measurements of turbulent fluxes of methane are conducted in the western part of the city centre (51°47'N, 19°28'E~~,~~) as shown in Fig. 1~~), in an area with~~). This part of the city has the highest population density~~, reaching~~ which reaches 17.2 thousand ~~persons~~people per km^2. The station for measurements of fluxes of mass, energy and momentum has been operating in the western part of Łódź since 2000 (Offerle et al., 2006a; 2006b, Pawlak et al., 2011; Fortuniak et al., 2013, Fortuniak and Pawlak, 2014), however, methane fluxes have been studied since July 2013. The measurement set is mounted on top of a mast at a height of z = 37 m (Fig. 2, left) which given the average height of buildings of 11 m, enables the assumption that the measurements are taken above the blending height in the inertial sub-layer (Fig. 2). The source area of turbulent fluxes was estimated (Fig. 1) for data collected during unstable stratification ($(z-z_d)/L < -0.05$) around midday (10.00–14.00) following the method of Schmid (1994) and ranged from 250 to 750 m away from the measurement station (Fig. 1).

The percentage of artificial ~~surfaces (~~surface coverage from buildings, ~~sidewalks~~pavements, streets~~,~~ and squares~~, etc.)~~ in this part of ~~town reaches~~the city is 62~~%, the~~%. The remaining part ~~being~~of the area is covered ~~with~~in vegetation~~,~~ of ~~which~~ only 10% ~~are~~is by trees (Kłysik, 1998). The vegetation is distributed unevenly in the form of lawns and trees growing along ~~the~~ street canyons. In the immediate vicinity of the measurement ~~point~~location, 3-5 storey ~~15-20 m high~~ buildings which range from 15 to 20 m in height dominate~~, built mostly in the 20th century.~~. Most of ~~them are characterised by~~the buildings have flat roofs covered with black tar paper or sheet metal.~~ The density of built-up areas north and east of the measurement point compared to the southern and western sectors is 10-20% greater (Fig. 1).~~ The trees growing in the area are mostly deciduous and their height usually does not exceed the height of the buildings~~, which results~~. Thisresults in a well-formed roof surface with an average height of 11 m. The ~~height~~density of built-up areas north and east of the measurement point, compared to the southern and western sectors, is 10-20% greater (Fig. 1). The displacement height z_d is estimated at 7.7 m. According to the classification by Stewart and Oke (2012), the local climate zone can be described as compact low rise. ~~The centre is surrounded by industrial and residential areas with tall 10-12 storey buildings or loosely built-up with single-family houses.~~ The roughness coefficient z_{0m} estimated for the neutral stratification surrounding the measurement point was ~~∼~~2.5 m on average. More information on the city's structure and the local climate conditions can be found~~, e.g.~~ in Kłysik (1996), Kłysik and Fortuniak (1999), Fortuniak et al. (2006, 2013), Offerle, (2006a, 2006b), Pawlak et al. (2011) and Zieliński et al. (2013). The gas distribution network and sewerage system around the flux tower are shown in Fig. 1.

~~The station for measurements of turbulent exchange of mass, energy and momentum has been operating in the western part of Łódź since 2000 (Offerle et al., 2006a; 2006b, Pawlak et al., 2011; Fortuniak et al., 2013, Fortuniak and Pawlak, 2014), but methane fluxes have been studied since July 2013, when the existing station for measuring sensible heat flux as well as water vapour and carbon dioxide was equipped with a fast response sensor for methane concentration fluctuations in the atmospheric air. The measurement kit is mounted on top of a mast at a height of z = 37 m (Fig. 2, left), which, given the average height of buildings of 11 m, enables the assumption that the measurements are taken above the blending height in the inertial sub-layer (Fig. 2). Based on the data obtained during the study period, the source area of turbulent fluxes was estimated (Fig. 1). To this end, Schmid's method (Schmid, 1994) was used, and all available data were used for the analysis. The analysis was performed for unstable stratification conditions~~ ~~($(z - z_d)/L < -0.05$)~~ ~~at the noon hours (from 10 a.m. to 2 p.m.). The significant height at which the measurement sensors were installed resulted in a large area of the source fluxes, which, depending on the wind direction, ranged from 250 to 750 m away from the measurement station during the study period (Fig. 1). The investigated sector of the city centre is a dense network of street canyons made available for motor traffic, i.e. one of the most important sources of greenhouse gases to the troposphere. The combustion of fossil fuels in vehicle engines produces water vapour, carbon dioxide and methane (when combustion is incomplete), which may also come from another source, i.e. leakage from vehicle natural gas tanks. Moreover, the measurement point is surrounded by a dense natural gas pipeline distribution network whose leaks lead to methane emissions into the troposphere (Fig. 1, bottom left). The dense sewerage system is another source of methane (Fig. 2, bottom, right).~~

2.2 Instrumentation and data processing

~~The measurements~~Measurements of~~ the~~ turbulent fluxes of methane were carried out using a standard measurement ~~kit. Its main unit was an ~~set consisting of the ultrasonic anemometer ~~RM Young~~RMYoung model 81000 (RMYoung, Traverse City, Michigan, USA~~), enabling the measurement of vertical wind velocity fluctuations. The station was equipped with ~~) and a fast response methane ~~fluctuation~~concentration sensor with an open measurement path ~~LI 7700~~LI7700 (Li-cor, Lincoln, Nebraska, USA). The measurements were carried out with a precision of 0.001 m·s^{-1} and 1 ppb respectively. As the final calculation of methane flux also requires ~~the ~~values of sensible heat ~~flux ~~and water vapour fluxes in the place of observation (LI 7700 instruction manual), the measurement ~~kit~~set also included a sensor ~~of~~measuring the ~~fluctuations~~concentrations of water vapour and carbon dioxide~~ LI 7500~~. This was a LI7500 Infra-Red CO_2/H_2O open path analyser (Li-cor, Lincoln, Nebraska, USA). ~~The fluctuations of air temperature necessary for the calculation of sensible heat flux were measured using the aforementioned ultrasonic anemometer RM Young.~~

The whole measurement system was attached ~~ca~~approximately 1 m below the top of the mast (Fig. 2, middle). ~~On~~The LI7500 head was placed on the horizontal arm~~,~~ on the south-eastern side of the mast at a distance of about 60 cm from the mast~~, the LI 7500 head was placed, and the~~. The ultrasonic anemometer was then installed at a distance of 20 cm. The ~~LI 7700~~LI7700 methane sensor was installed on an additional arm, ~~slightly~~30 cm lower, so that the centre of its measurement path, which is about ~~4~~four times longer than the paths of ~~LI 7500~~LI7500 and ultrasonic anemometer, was at a similar level. ~~As follows from earlier analyses,~~Previous studies have shown that the influence of ~~the~~a mast ~~whose~~of diameter ~~is about ~~0.15 m is negligible and does not generate ~~a~~ flow distortion (~~for details, see ~~Fortuniak et al., 2013).

All ~~of ~~the aforementioned sensors ~~measured the fluctuations of parameters~~sampled with a frequency of 10 Hz. Immediately before starting the measurements in July 2013, the sensor for measuring H_2O and CO_2 ~~fluctuations~~mole fractions was calibrated (the zero and span values were set). The methane concentration analyser was installed directly after purchase, so the zero and span had been set by the manufacturer. The two sensors and the ultrasonic anemometer were cleaned approximately once a month~~, with~~. This was pertinent to the methane sensor ~~needing it in the first place, as~~because its mirrors proved to be highly susceptible to grime (air impurities, bird droppings, atmospheric deposits, drying raindrops or melting snowflakes). ~~Although the~~The manufacturer ~~had ~~equipped the instrument with a

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mirror heating and condensation anti-freezing system ~~as well as~~and a cleaning system~~ which, by means of~~. Using a pump, this applied ~~the ~~cleaning liquid to the lower mirror~~;~~. However, in practice~~, however, especially~~ and particularly in autumn and winter~~,~~ this ~~turned out to be~~was insufficient~~. There were situations, for example~~ particularly on days with humidity of up to 100~~%,~~% when the signal strength dropped by several tens of percent in just a few hours. According to the manufacturer of the instrument, if the signal strength ~~(RSSI –~~Relative Signal Strength Indicator (RSSI) is less than 10%, this means that the measurement path ~~was~~is blocked by external factors. ~~During the measurements, however~~However, it was decided to tighten this criterion~~, and in~~. In order to calculate the fluxes, the methane ~~fluctuation~~mole fraction values observed at RSSI >20% were chosen. ~~The signal strength of the instrument when measuring the concentration of methane only in about 8% of cases~~ Of these, the RSSI exceeded 70% in only 8% of cases (Fig. 2, right), while observations at 20~~%<~~% < RSSI~~<~~ < 70% had a much greater share~~, and most~~. Most often~~,~~ and in 20% of cases~~,~~ the signal strength ~~value~~ was between 30% and 40% (Fig. 2, right).

The 10 Hz fluctuation data for the vertical wind velocity and the concentrations of water vapour and methane were recorded by a CR21X datalogger (Campbell Scientific, Logan, Utah, USA) so that all parameters could be recorded at the same time. The measurement station was also equipped with sensors recording the general weather conditions (air temperature and humidity, atmospheric pressure, wind direction and velocity, radiation balance components, precipitation). These data were recorded every 10 minutes by a CR10 datalogger (Campbell Scientific, Logan, Utah, USA) and were archived together with ~~fast changing~~the 10 Hz data ~~archived~~ on a PC. ~~The turbulent fluxes of methane were calculated using software written by the authors in Fortran 77.~~

The ~~methane flux (~~FCH_4~~)~~ was determined directly from the definition as the ~~covariability~~covariance of the vertical wind velocity fluctuations and the methane concentration fluctuations in the air (Lee et al., 2005; Foken, 2008; Burba and Anderson, 2010; Aubinet et al., 2012):

$$\cancel{FCH_4 = \overline{w'\rho CH_4'} = \frac{1}{N}\sum_{N}^{i=1}\left(w' - \overline{w}\right)\left(\rho CH_4' - \overline{\rho CH_4}\right).}$$

$$FCH_4 = \overline{w'\rho CH_4'} = \frac{1}{N}\sum_{N}^{i=1}\left(w - \overline{w}\right)\left(\rho CH_4 - \overline{\rho CH_4}\right). \qquad (1)$$

Kod pola został zmieniony

The *w'* and *ρCH4'* parameters are, respectively, the fluctuations of vertical wind velocity and the concentration of methane in the air, while $\bar{w}$ and $\overline{\rho CH_4}$ are their averaged values. A positive flux means the turbulent transport of methane into the troposphere, a negative flux is its ~~intake~~uptake by the urban surface. ~~In the calculations,~~ 1 hour block averaging was used~~, with 1 hour being used~~ as an averaging period. Since the measurements were carried out at a considerable height, a shorter averaging period could lead to underestimating the fluxes (Pawlak et al., 2011). During the calculations, all necessary procedures and corrections were applied. Any data with non-real values were rejected, the spike detection procedure was performed (Vickers and Mahrt, 1997), the double rotation of the wind coordinate system was applied (Kaimal and Finnigan, 1994) and the impact of separation of the sensors was eliminated by maximizing the ~~covariability~~covariance in the interval +/-2 s. Furthermore, sonic temperature was corrected for humidity in the air (Schotanus et al., 1983) and the WPL correction was added (Webb et al., 1980). According to ~~LI-7700~~LI7700 manufacturer's recommendations, the correction terms related to air density fluctuations affecting both the spectroscopic response and the mass density retrieval were applied ~~(LI-7700~~ (LI7700 instruction manual).

A detailed control of the quality of the calculated fluxes was also carried out~~,~~ which focused primarily on the assessment of data stationarity. The most commonly used Foken's test (Foken and Wichura, 1996) is not always fit for ~~the~~ purpose~~, and therefore~~. Therefore two other tests were used~~,~~ as proposed by Mahrt (1998) and Dutaur et al. (1999~~),~~,) modified by Affre et al~~.~~., (2000). During the data quality assessment, a very strict criterion was adopted~~, which classified~~ to classify the data as suitable for further analysis when ~~and only when all the~~ three tests confirmed that ~~the condition of~~ stationarity was met. A milder criterion, indicating good data quality if at least one test suggested stationarity~~,~~ did not meet the expectations ~~because it~~. This criterion accepted ~~a number of~~ data with unrealistically high positive values and a substantial number of fluxes with high negative values whose existence ~~is physically difficult to explain. On the one hand~~cannot be explained. However, the restrictive evaluation of the data reduced the ~~number~~amount of data suitable for further analysis by 23.8~~%, but on the other hand, uncertainty~~%. Uncertainty regarding their quality was kept to a minimum. ~~Earlier, about~~Approximately 10% of the data were not registered due to problems with ~~electricity and the computer~~power supply in autumn 2013, and 29.8% of the recorded data were rejected because the measurements had been taken in weather conditions which made it impossible for the ~~LI 7700~~LI7700 sensor to measure the concentration of methane properly ~~(~~. This was a result of such factors as precipitation and atmospheric deposits, saturation of air with water vapour~~,~~ and impurities~~, etc.)~~. This problem ~~used to occur~~occurred particularly in autumn and winter (Table. 1) when ~~frequently~~frequent cleaning of the sensor placed on the mast was impossible. As a result~~,~~ the percentage of ~~good~~acceptable data was 36.4% ~~(~~as shown in Table. 1~~).~~ .

3 Results

3.1 Climate background

The climate of central Poland ~~where Łódź is situated~~ is a typical transitional climate of moderate latitudes~~, formed~~. It is characterised by marine air masses flowing from the west and by continental air from the east. The mean monthly air temperature in the study period varied from 0.1°C in winter (January 2014) to 22.8°C in summer (August 2015). The ~~years were~~study period was considered to be hot~~,~~ with ~~heatwaves~~heat waves occurring~~,~~ and the winters were relatively warm with mean temperatures in 2014 and 2015 of 2.7°C and 2.4°C~~,~~ respectively. The average temperature in 2014 was 10.9°C. The ~~average~~ total precipitation in the same year was 584 mm, with a greater amount of precipitation of 360 mm (61.6% of the annual total) recorded in the warm half~~-~~ of the year. The maximum solar radiation ~~totals were~~was observed in July (688 MJ·month^{-1} in 2014 and 697 MJ·month^{-1} in 2015~~),~~) while the minimum ~~totals~~ occurred in the winter months when they fell below ~~100~~80 MJ·month^{-1}. The monthly radiation balance totals were almost 400 MJ·month^{-1} in July, while in the winter months they became negative and reached even -56 MJ·month^{-1} (December 2013). The average wind speed in the period was 3.1 m·s^{-1}, with slightly higher values in winter (3.4 m·s^{-1}) and lower values in the summer (2.8 m·s^{-1} on average). The study area of the city is dominated by air flow from the west (~~for details, see~~ Fortuniak et al., 2013). ~~Generally, it can be noted that during~~

During the measurement period, atmospheric instability or ~~neutrality~~neutral conditions prevailed in the city centre. ~~A stable~~Stable air stratification ~~could rarely be~~was observed in the centre of Łódź~~,~~ in only ~~in~~ 7.6% of cases (from 2.9% in winter to 10.4% in summer). The frequency of neutral and unstable stratification was ~~very~~ similar and was~~, respectively,~~ 46.0% and 46.4~~%. Instability~~% respectively. Unstable conditions prevailed in summer (51.6% of cases), while ~~neutrality was~~neutral conditions were observed in 61.7% of cases in winter. In the diurnal cycle, stable stratification was also a rarity. In the daytime (10.00 a.m. to 2.00 p.m.), this type of stratification was observed in only 0.3% of cases on average throughout the year, while at night the condition ξ> 0 was met by 15.0% of the data. Other types of atmospheric ~~equilibrium~~stability appeared in the daytime in 19.7% (neutral) and in 80% (unstable) of cases. At night, ~~atmospheric neutrality~~neutral conditions prevailed (67.0% of cases), while ~~instability was~~unstable conditions were observed in 18.0% of cases on average throughout the year.

Clear annual and diurnal cycles characterized the fluxes of energy and mass. Both the sensible heat flux Q_H and the latent heat flux Q_E were largest in summer (about 190 MJ·month^{-1}and 120-150 MJ·month^{-1}, respectively~~).~~.) The ~~values of the~~ Bowen ratio $B=Q_H/Q_E$ ~~were~~was typically urban~~,~~ i.e. greater than 1 ~~(~~and up to 2.25 in May 2015). The annual variability of carbon dioxide flux (FCO_2) was also marked by an annual cycle. The maximum values occurred in winter when anthropogenic CO_2 emissions, being a result of burning fossil fuels ~~(~~for vehicle traffic~~,~~ and domestic heating~~, etc.),~~ were the largest. The typical values exceeded 20 μmol·m^{-2}·s^{-1} and had a maximum of ~55 μmol·m^{-2}·s^{-1}. In summer, ~~when~~ the consumption of ~~carbon dioxide~~CO_2 by urban vegetation and ~~reduced emissions of this gas due to the~~ lack of ~~need to heat homes~~domestic house heating contribute to a decrease in the intensity of net exchange~~, the~~. The minimum values of FCO_2 were observed~~, from~~ as -10 to 10 μmol·m^{-2}·s^{-1}.

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3.2 Annual variability of FCH_4

The two-year measurements of ~~turbulent methane flux (~~FCH_4~~)~~ revealed a number of characteristics of the exchange of methane in the city-troposphere system. ~~First of all, regardless~~Irrespective of the season~~, there was a definite domination of~~ mainly positive ~~values~~ of FCH_4 ~~(~~were observed (Fig. 3). On average, the percentage of positive values over the study period was 93.7~~%, being~~% which was slightly greater in the cold season (94.6%) than in the warm season (93.2~~% of cases).~~%). This means that~~,~~ regardless of the season~~,~~ the centre of Łódź is a source of methane to the atmosphere. In addition, the time variability of FCH_4 shows a clear annual cycle with a maximum in the cold season and a minimum in the warm season (Fig. 3). The highest recorded values ~~clearly~~ exceeded 100 nmol·m^{-2}·s^{-1} and were observed in November, December, January and February. The least intense exchange of methane was observed from May to September, when FCH_4 was rarely greater than 50 $nmol \cdot m^{-2} \cdot s^{-1}$. The exception was the summer of 2013 when the recorded values of FCH_4 were close to winter values in July and August. However, only average values were elevated, while the median values are similar to those of July and August 2014 and 2015. It can be assumed that in the summer of 2013 additional sources of methane were present which could be the result of damages to the gas network. It is likely that this occurred south-east of the station where the deep excavations associated with the construction of a tunnel for one of the main streets of the city centre was completed (Fig. 2).

It seems that the annual cycle of turbulent methane exchange should be attributed to the anthropogenic origin of this gas in the centre of the city. In the cold season, there ~~occurs~~is an increase in methane emissions associated with the combustion of fossil fuels, which results from the increased discharge of motor vehicle exhaust gas~~, being a significant source of methane in the city.~~ (Heeb et al., 2003; Nakagawa et al., 2005). Another important factor is the increased natural gas consumption in winter, its leakage from distribution networks and ~~during the~~its use ~~of~~in domestic gas burners. ~~Certain amounts of methane are~~Methane is also produced by heating ovens (Ciais et al., 2013). The absence of inventory data makes it difficult to verify these dependencies ~~in the case of~~for Łódź~~; however~~. However, the increased values of the flux of methane are clearly visible ~~in the periods of~~where there are rapid drops in air temperature~~,~~ i.e.~~g.~~ in late October or late November and December 2014 (Fig. 3). A pronounced annual cycle can also be seen in the ~~time~~temporal variability of the mean monthly values of FCH_4 (Fig. 3, Table 2). The highest monthly averages of FCH_4 were recorded in January and February 2014~~,~~ when the average exchange exceeded 60 $nmol \cdot m^{-2} \cdot s^{-1}$. In the same months of 2015, the FCH_4 values were lower and slightly exceeded 50 $nmol \cdot m^{-2} \cdot s^{-1}$, which was a consequence of winter 2014/2015 being warmer ~~as compared to the previous one.~~than 2013/2014. The mean monthly values of FCH_4 in summer rarely exceeded 20 $nmol \cdot m^{-2} \cdot s^{-1}$. The median values in the warm half of the year were very similar to the average values. In the cold season, the median was lower due to the sporadically occurring elevated levels of FCH_4. Regardless of the ~~measurement~~measurements, some differences in the time variability of methane flux in transitional seasons can also be observed. In late winter and early spring, a rapid drop in FCH_4 by ~~about~~approximately 30 $nmol \cdot m^{-2} \cdot s^{-1}$ can be observed, while FCH_4 starts to increase at the end of summer and slowly continues until winter. The cold half of the year is also characterised by a greater variability of the ~~turbulent exchange~~fluxes of methane (Table ~~3~~2). In the summer months, the standard deviation of

FCH_4 did not exceed 20 nmol·m^{-2}·s^{-1}, whereas during the winter months it was more than ~~twice~~two times greater. An exception is the aforementioned summer of 2013.

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3.3 Diurnal variability of FCH_4

~~Figure~~Fig. 4 shows the average daily flux of methane in the centre of Łódź calculated for the entire study period (top graph) and for the successive months of the year (middle and bottom graphs). ~~Most importantly, the~~The average daily variability in the successive months confirms the above described annual variability~~:~~ i.e. higher values of FCH_4 occurred in the cold season. Furthermore, the average daily variability, regardless of month and time of day, is always positive~~, which~~. This means that the emissions of methane ~~definitely~~ dominate over its uptake by the urban surface. ~~In the case of the~~The daily pattern, averaged for the entire measurement period, ~~it certainly represents~~show a clear diurnal cycle with two maxima and two minima. The maximum values occurred in the morning (7.00 - 8.00 UTC + 1) and in the ~~afternoon~~evening (19.00 - 20.00 UTC + 1). During the maxima, the values of FCH_4 reached almost 40 nmol·m^{-2}·s^{-1}, whereas during the noon hours and at night they dropped to 26-28 nmol·m^{-2}·s^{-1}. Such a daily pattern suggests that the average flux of methane can be divided into two components. One has an approximately constant value of up to 26-28 nmol·m^{-2}·s^{-1}~~,~~ and its source may be the sewerage system ~~or~~and the natural gas distribution system. ~~Before noon~~In the morning and in the afternoon, additional sources of methane (vehicle traffic, combustion of natural gas, leaks from gas network~~,~~ associated with the ~~distribution~~ increasing ~~during the day~~gas consumption) are ~~activated~~activate, increasing the flux by 10-12 nmol·m^{-2}·s^{-1}. ~~Due~~However, it should be noted that due to the lack of inventory data, the above considerations are only hypothetical. ~~In the following months, the average daily variability was not so clear.~~

In the warm half of the year (May-October), the average daily variability was low and from April to September it ranged between 10 and 40 nmol·m^{-2}·s^{-1}. In May, June and July~~,~~ it was difficult to see clear maxima during a 24~~ hours~~ hour period. In August, September and October there was a maximum in the morning. In the cold half of the year (November-April), the average daily variability of FCH_4 was characterised by distinctly higher values~~,~~ from 20 to 90 nmol·m^{-2}·s^{-1}. ~~At that time~~In this period, the double daily maximum was easier to identify~~,~~ and in November, December, January and February the afternoon peak seemed to be ~~by a few~~

~~nmol·m^{-2}·s^{-1}~~ greater than in the mornings. In March and April, the maximum values were comparable. The presence of two maxima in the variability of FCH_4 in the cold season could be explained by the increased consumption of natural gas, the combustion of fossil fuels in the morning and afternoon hours ~~(~~from cooking~~,~~ and domestic heating ~~homes)~~ and the diurnal variability of motor vehicle traffic~~,~~ which ~~is often accompanied by traffic jams~~can cause road congestion in winter. In the ~~warm season of the year (and especially~~warmer seasons and particularly during the ~~holiday~~holidays periods~~)~~,, motor vehicle traffic became less intense and the city's inhabitants stopped heating their homes. In the cold season, FCH_4 is also characterised by greater variability throughout the day. The standard deviation of FCH_4 in this season ~~is even up to~~can reach 50 nmol·m^{-2}·s^{-1}~~,~~ while in the warm season it rarely exceeds 20 nmol·m^{-2}·s^{-1}.

Sformatowano: Indeks dolny

3.4 Monthly and annual exchange of FCH_4

Based on the average daily patterns of FCH_4 calculated for each month~~,~~ (the sum of the average hourly FCH_4 multiplied by the number of days in the month), the exchange of methane in the successive months of the study period was determined (Fig. 5). The highest values occurred in winter ~~when they were up to 2.0 g·m^{-2}·month^{-1}, and~~where in January and February 2014 they exceeded 2.5 g·m^{-2}·month^{-1}. The summer values were more than ~~twice~~two times lower and dropped to 0.7-0.8 g·m^{-2}·month^{-1}. The autumn of 2013 was ~~characterised~~characterized by elevated values of FCH_4. A comparison between the monthly exchange of methane and the mean monthly air temperature reveals a clear link between these parameters (coefficient of determination = 0.731, Fig. 5, bottom right). ~~Therefore~~So, the anthropogenic sources of methane gain intensity at low air temperatures, which can be seen by comparing the results of FCH_4 measurements in winter 2013/14 and 2014/15 (Fig. 5). In the first case, an increase of the monthly values of the flux was recorded starting from November, with a maximum in January and then a decrease until April-May. Between November 2013 and January 2014, the exchange almost doubled (from 1.36 to 2.67 g·m^{-2}·month^{-1}). The next winter, the monthly exchange of methane between November and March differed ~~only slightly~~little, and FCH_4 increased from November 2014 to January 2015 only by ca 0.27 g·m^{-2}·month^{-1}. The differences were associated with thermal contrasts during the two winters. In winter 2014/2015, the monthly average temperature remained at 2.2-2.5°C, while ~~the mean January temperature~~ in the previous winter season the mean January temperature dropped to 0.1°C. The greater activity of the anthropogenic sources of methane in the centre of the city is also confirmed by the measurements of methane concentrations (Fig. 5, bottom left). The high winter values of the flux of methane are accompanied by higher concentrations of the gas in the air~~.~~ and seasonal changes in OH concentration.

Based on the data on the exchange of methane in Łódź obtained between January 2014 and August 2015, an attempt was made to assess the cumulative annual exchange of this gas in the centre of Łódź between the city centre and the atmosphere (Fig. 6). To date, there have been no standard methods for ~~supplementing~~filling gaps in the long-term data series of turbulent fluxes of methane in ~~urbanised~~urbanized areas. Difficulties with their development arise primarily from the fact that the continuous measurements of FCH_4 in cities are still rare. Furthermore, as in the case of carbon dioxide fluxes, data on anthropogenic sources of the gas and the parameters of natural processes (e.g. air temperature) ~~are required~~may be useful for the data gap filling procedures (Aubinet et al., 2012). The annual exchange of methane in the city centre was therefore estimated using two simple methods. Firstly, on the basis of the average daily patterns of FCH_4, the monthly exchange in the successive months was determined and then the accumulation was made (Fig. 6, solid step plots). Secondly, the gaps were filled in a series of 1-hour values of FCH_4 in two ways. If a data gap was not longer than 3 hours, interpolation was used, while for longer gaps~~,~~ data were inserted from the average daily pattern in the respective month for the respective hour. Both methods yielded very similar results (the difference was ~~~~~approximately 1%), although it is obvious that the cumulative fluxes obtained in this manner should be regarded as an approximation. Therefore~~,~~ it can be stated that the annual exchange of methane in the centre of Łódź in 2014 was equal to about 17.6 $g{\cdot}m^{-2}$ (Fig. 6). The graph shows the impact of the annual variability of methane flux: the cumulative flux grows fastest in the cold half of the year. Due to the differences in the exchange of methane described in ~~section~~Section 3.4, ~~from the point of view of~~with reference to changes in air temperature in the study period, the cumulative exchange in the period January-August 2015 was calculated in a similar manner. The relatively warmer beginning of 2015 caused the exchange to be less intense and the cumulative flux of FCH_4 in August 2015 was by 9.2% lower than in 2015.

3.5 Weekly differences of FCH_4

Since ~~the turbulent exchange~~fluxes of methane in the city ~~is~~are associated with anthropogenic sources~~ of this gas~~, a weekly cycle of FCH_4 should be expected~~,~~, which is similar ~~that~~to that seen ~~in the case of carbon dioxide~~for CO_2 exchange (Pawlak et al., 2011). Based the on 1-hour data for FCH_4 recorded in the period~~ from~~ July 2013 to August 2015, an average daily flux of 44.3 $mg \cdot m^{-2} \cdot day^{-1}$ was determined (Fig. 7). Having taken only working days for the calculation (Monday to Friday), it was found that the exchange was higher~~,~~ i.e. ~~it was~~ 45.2 $mg \cdot m^{-2} \cdot day^{-1}$. ~~On the other hand~~In contrast, the average daily exchange of methane during weekends (Saturday and Sunday) amounted to 42.3 $mg \cdot m^{-2} \cdot day^{-1}$ and was therefore lower by 4.5%. ~~Thus, it can be concluded~~These results suggest that, on average, in the study period~~,~~ anthropogenic sources of methane ~~were~~ are likely to be less ~~active~~ at weekends compared to working days. It should be emphasized that on Saturday, the average flux was lower by 6.9% in comparison with working days~~.~~, while on Sunday it was lower by 12%. The difference is a result of significantly lower peak on Sunday morning, which can be attributed to less intensive human activity on Sunday morning and lower traffic load in comparison to the same time of day on Saturday. Similar results were observed in summer and winter, when the average daily exchange on working days was higher by 1.6% (summer) and 1.9% (winter) ~~$mg \cdot m^{-2} \cdot day^{-1}$ when~~ compared with the average for the whole week. The average daily exchange at weekends was~~, in turn,~~ lower by 4.0% and 4.7% respectively~~ 4.0% and 4.7%.~~. An exception is the transitional seasons when the average daily exchange of methane on working days was comparable (spring, -0.3%) or slightly lower (autumn, -1.6%) than the average for the entire week (Fig. 7). On the other hand, the average daily exchange during the weekend turned out to be higher and amounted to +1.6% (spring) and +3.8% (autumn). Without the inventory data, it is difficult to explain ~~such values of~~why the fluxes~~, especially because~~ vary, particularly in the case of ~~carbon dioxide~~CO_2 fluxes~~,~~ where higher values are observed on working days as compared to weekends throughout the year (Pawlak et al., 2011).

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3.6 Methane ~~turbulent exchange~~fluxes and wind direction

~~The~~As mentioned in Section 2.1, the centre of Łódź is the most densely built-up area of the city. The measurement point is located in an area of uniform ~~similar~~ building density ~~parameters,~~ while, as mentioned in ~~section~~Section 2.1, this density is slightly greater to the east and north of the station. An analysis of the average value of ~~-~~FCH_4 depending on the wind direction confirms, at least in part, the impact of building density on the value of methane turbulent exchange (Fig. 8). The fluxes of ~~CH_4~~methane recorded during airflow from the north, and especially from the south-east, were by far the largest in the study period and reached 35-45 nmol·m^{-2}·s^{-1} (Fig. 8, left). However, it is difficult to ~~regard~~ be confident in the direct relationship between urban design~~-~~ and FCH_4 ~~as certain, due to~~because of the increased values of FCH_4 ~~coming also~~ from the south-western sector (approximately 40 nmol·m^{-2}·s^{-1}). ~~Of course, such~~Such a relationship cannot be ruled out~~;~~, however~~,~~ the local point sources of methane may play an important role~~, but~~ even though they are difficult to identify. In the case of the south-western sector, the liquid petroleum gas (LPG) station located approximately 800 m from the measurement station may be such a source. It lies approximately 200 m to the west of the large intersection where traffic load is usually larger than the surrounding streets. Significantly lower values of FCH_4 (less than 20 nmol·m^{-2}·s^{-1}) observed with airflow from the south and west may be due to the presence of large urban parks (Fig. 1).Heavy traffic does not run through these streets and the density of the gas network and sewage system is also significantly smaller in comparison with other sectors (Fig. 1). The distribution of average ~~values of~~ FCH_4 depending on the wind direction, calculated for the cold half of the year (Fig. 8, middle~~),~~) suggests that in this season~~ of the year the~~ local anthropogenic methane sources were more intense. The relationship between FCH_4 and the wind direction was much the same throughout the study period, while the average values of fluxes were higher and amounted to 55-70 nmol·m^{-2}·s^{-1}. Therefore, the sources could be~~, e.g.,~~ clusters of houses with leaks from gas installations~~,~~ or vehicles at nearby intersections~~,~~ which are heavily jammed in the cold half of the year. In summer, the average fluxes of CH_4~~, regardless of the wind direction,~~ were significantly lower (less than 30 nmol·m^{-2}·s^{-1}, Fig. 8, right~~), and contrasts~~) regardless of the wind direction. The contrast between the sectors ~~were~~was not ~~too~~ clear. An exception is the clearly visible elevated value of FCH_4, associated with the airflow from the south-western sector.

3.7 Methane ~~turbulent exchange~~fluxes in relation with carbon dioxide fluxes

~~The measurements of turbulent mass fluxes in urban areas are performed relatively rarely (Aubinet et al., 2012). This applies to the fluxes of carbon dioxide, but in the first place to the fluxes of methane. While the measurements of FCO_2 in cities have been carried out for more than a decade and the results summarising such long-term measurements in at least a dozen cities around the world have been published to date, similar results concerning the fluxes of methane are still lacking.~~ During more than two years of measurements in Łódź, both FCO_2 and FCH_4 were measured and therefore the question about ~~the~~ temporal covariability of both fluxes ~~should~~can be ~~asked~~addressed, and, consequently, whether the exchange of methane can be estimated based on the knowledge of the flux of CO_2. ~~To this end, the~~The average daily variability (Fig. 9, left) and the average monthly variability (Fig. 9, middle) of the value of methane flux were compared to the fluxes of CO_2. As the figure indicates, such covariability exists ~~-~~and bigger fluxes of CH_4 are accompanied by larger fluxes of CO_2. Unfortunately, the ~~values of the coefficient~~low coefficients of determination ~~of~~ (0.57 and 0.56~~, which were not very high, leave no illusions about the possibility of using~~)mean FCO_2 cannot be used as a proxy for FCH_4 in the centre of Łódź. An even weaker relationship was observed between the average daily patterns of FCH_4 and FCO_2 (Fig. 9, right). Although the two fluxes have a characteristic pattern with two maxima in a 24-hour period, the coefficient of determination is only 0.25. ~~It is~~We therefore ~~difficult~~conclude that FCO_2 data cannot be used to ~~talk about covariability~~ facilitate gap-filling of ~~the fluxes of CO_2 and CH_4~~FCH_4 data in the centre of Łódź~~, which could, for example, facilitate the process of filling gaps in the series of methane fluxes based on the FCO_2 data.~~ .

The comparison of FCH_4 and FCO_2 fluxes allow analysis of the relative contribution of each of the fluxes to total emissions to atmosphere. The average value of the FCH_4/FCO_2 ratio in 2013-2015 was $3.71*10^{-3}$ (Fig. 10, top). Rather stable values of the ratio in months (minimum $2.41*10^{-3}$, maximum $5.3*10^{-3}$) and the lack of a clear annual course suggest rather comparable magnitude of both fluxes. However, a clear diurnal course in the ratio has been observed (Fig. 10, bottom) with reduced values in the day and elevated values at night. On average, over the study period and in the transitional seasons, the daily variation of FCH_4/FCO_2 was similar. Between the hours of 9 a.m. and 5 p.m. FCH_4/FCO_2 was approximately constant of the order 2.5 to $3.5*10^{-3}$. At night, these values grow to about $5\text{-}7*10^{-3}$ which can be explained by relatively constant methane emissions related to leaks from pipelines, and reduced emission of CO_2 which is the result of minimum of traffic load. In winter, the average daily variability of the FCH_4/FCO_2 ratio can be characterised by slightly higher values during the day (about $4.4*10^{-3}$) and significantly higher at night reaching $12*10^{-3}$ between the hours of 2 a.m. and 6 a.m. (Fig. 10, bottom). The causes again are clear: a minimum traffic load giving reduced fluxes of FCO_2 but also increased methane leaks from pipelines associated with higher gas consumption for heating of the surrounding buildings. The exception was the daily course FCH_4/FCO_2 in the summer which is reversed. The minimum (of the order of $3\text{-}5*10^{-3}$) was observed at night and the maximum (more than $8*10^{-3}$) around noon (Fig. 10, bottom). Elevated values of the ratio during the day are the result photosynthesis reducing FCO_2 flux.

4 Summary and conclusions

The measurements of the methane flux (FCH_4) carried out in the centre of Łódź for more than two years provided information on the time variability of ~~turbulent exchanges of~~ methane exchange between the urban surface and the atmosphere. The measurement results showed that, as in the case of other greenhouse gases, i.e. water vapour (Offerle et al., 2006a, 2006b) and ~~carbon dioxide~~CO_2 (Pawlak et al., 2011), the centre of Łódź is a source of methane to the atmosphere. Another feature indicating the similarity in the time variability of greenhouse gases is the annual cycle of the exchange of methane in the system: ~~the~~ city centre ~~to the~~ /atmosphere, which seems to result from an annual cycle of anthropogenic methane emissions. Other characteristics such as diurnal variability, and notably weekly variability, are not as pronounced as ~~in the case~~for FCO_2 (Pawlak et al., 2011). The annual exchange of methane in terms of pure carbon in the centre of Łódź was estimated at 13.2 $gC\cdot m^{-2}\cdot year^{-1}$, which, compared to the exchange of ~~carbon dioxide~~CO_2 estimated in Łódź at 2.93 $kgC\cdot m^{-2}\cdot year^{-1}$, does not seem too large ~~a quantity. However, it should be kept in mind that the greenhouse potential of methane is much higher, which is one reason why the measurements of this gas exchange should not be marginalised in favour of carbon dioxide.~~. At the same time, it must be noted that the centre of Łódź~~, in terms of intensity,~~ is a source of methane comparable in intensity to ~~natural areas considered to be~~ the most productive natural areas , i.e. wetlands. The annual exchange of methane in Łódź was estimated to be 17.6 $g\cdot m^{-2}\cdot year^{-1}$, while at the same time (2014) the exchange in the wetlands of the Biebrza National Park (north-eastern Poland) was approximately 18 $g\cdot m^{-2}\cdot year^{-1}$ (Fortuniak and Pawlak, 2015~~, paper~~

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~~in preparation).~~). Comparable values ~~for~~of the annual exchange of methane were also observed at other stations located in wetlands: ~~approx.~~approximately 16.5 g·m^{-2}·year^{-1} (Finland, Rinne et al., 2007~~),~~) or 14.0 - 18.5 g·m^{-2}·year^{-1} (Sweden, Nilsson et al., 2008).

Unfortunately, ~~currently, the~~ possibility of comparing the results obtained with those from other cities is ~~severely~~ limited~~.~~ at this time. The only longer-term measurements of ~~the~~ ~~CH_4~~methane flux were performed in Florence (March - May 2011, Gioli et al., 2012) and in London (~~summer 2012, O'Shea~~3-year campaign, Helfter et al., ~~2014~~2016). The mean values of the ~~CH_4~~methane fluxes obtained in these cities were higher than in Łódź. In Florence, the average methane exchange in the spring of 2011 was estimated to be 135 nmol·m^{-2}·s^{-1}~~, while the~~. The average FCH_4 in Łódź in the same season was ~~4~~four times lower~~,~~ and equal to 31 nmol·m^{-2}·s^{-1}. On the other hand, a comparison of the obtained results of FCH_4 measurements with inventory research does not necessarily yield a positive outcome (Gioli et al., 2012). In London, ~~in turn,~~ the average exchange ~~during the summer~~ was also several times higher than that observed in Łódź (~~140~~142 and ~~21~~32 nmol·m^{-2}·s^{-1}, respectively). The results of this type, however, allow only a very general comparison~~,~~ and the ~~brief periods of research prevent further~~limited sampling period prevents analysis. For example, the mean variability of daily FCH_4 in Florence in spring was characterised by one maximum during the day, while two maxima were observed in Łódź ~~at~~for the same ~~time: before~~period: morning and ~~after noon~~evening. The measurements in Florence showed no correlation between FCH_4 and air temperature (R^2=-0.04, Gioli et al., 2012), while in Łódź~~, such~~ a ~~relation~~strong relationship occurs (R^2=0.71). It is also impossible to compare the annual exchange~~,~~ and~~,~~ in the absence of measurements in other cities, it is difficult to determine the relationship between the intensity of annual methane exchange and a parameter ~~characterising~~characterizing the study area of the city in ~~a~~ general~~ way~~. In the case of ~~carbon dioxide~~CO_2 flux, a clear relationship between the annual FCO_2 and the percentage of artificial surfaces in the vicinity of the measurement point (Nordbo et al., 2012; Oliphant, 2012) was observed. Based on the existing measurements, it is difficult to attempt to seek a similar dependence for the flux of methane~~.~~ since only in London a relationship between FCH_4 and population has been found (Helfter et al., 2016). There are also several published results of urban methane emissions obtained using eddy-covariance techniques such as using alkanes (Los Angeles, Pieschl et al., 2013), aircraft measurements (Indianapolis, Mays et al., 2009) or a ground-based Fourier transform spectrometer (Los Angeles, Wunch et al., 2009). All of them reports existence of higher FCH_4 fluxes than measured in Łódź.~~On the other hand, a comparison of the obtained results of~~

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~~FCH_4 measurements with inventory research does not necessarily yield a positive outcome (Gioli et al., 2012).~~

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Table 1. Data capture of 1-hour values recorded for FCH_4 in the ~~centre~~center of Łódź in the period ~~July2013~~July 2013 – August 2015

Spring MAM	39.1%
Summer JJA	47.4%
Autumn SON	26.5%
Winter DJF	31.1%
July 2013 – August 2015	36.4%

Table 2. Monthly values of mean, median and standard deviation values of FCH_4 in the centre of Łódź in the period ~~July2013~~July 2013 – August 2015 (all fluxes in $nmol \cdot m^{-2} \cdot s^{-1}$).

		J	F	M	A	M	J	J	A	S	O	N	D
2013	Mean	-	-	-	-	-	-	22.1	29.5	39.6	38.1	35.3	45.3
2013	Median	-	-	-	-	-	-	18.2	22.0	26.3	27.7	26.6	38.6
2013	St. dev.	-	-	-	-	-	-	27.1	35.7	56.2	43.7	39.3	35.9
2014	Mean	62.9	66.6	37.4	33.1	21.8	22.9	20.3	19.2	20.8	27.0	43.4	47.8
2014	Median	60.2	64.4	30.6	31.7	20.8	22.1	19.3	18.2	20.2	23.6	34.2	38.4
2014	St. dev.	46.7	42.6	32.9	21.8	20.8	16.8	14.7	15.2	11.3	20.9	31.9	39.3
2015	Mean	52.5	54.2	48.6	25.2	22.8	18.4	17.6	21.5	-	-	-	-
2015	Median	47.6	51.8	46.5	22.4	22.1	17.7	17.3	20.3	-	-	-	-
2015	St. dev.	34.5	39.4	32.4	23.7	16.2	14.9	12.9	14.6	-	-	-	-


1 0 1 2 3 4 5 Kilometers

Figure 1. The western part of the ~~centre~~center of Łódź (top). Solid white lines indicate the source area with P = 25, 50, 75 and 90%, calculated for the turbulent fluxes measured at 10~~-14 hours~~ a.m. to 2 p.m. during unstable stratification (all available data from the period July 2013 - August 2015). The dashed red lines represent the 250, 500, 750 and 1000 m distance from the measurement point. White circle indicates LPG station and blue rectangle indicates area of road tunnel construction. Bottom figures show spatial distribution of gas network (bottom left) and sewage system (bottom right) in the neighbourhood of the measurement site (white dots). Schemes are based on data from Geodesy ~~Centre~~Center of Łódź (www.mapa.lodz.pl). Photo source: www.google.com

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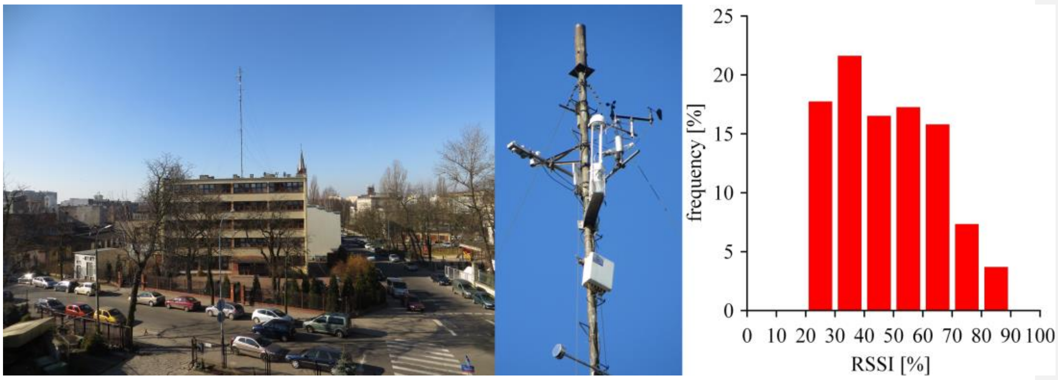

Figure 2. A FCH_4 measurement site in Łódź (left) and instrumentation ~~set~~ (middle). ~~Right~~The right figure shows the frequency of measured 1-hour blocks of raw data in relation ~~with~~to the RSSI (Received Signal Strength Indicator) of Li7700 methane open path ~~analyzer~~analyser. Data recorded only in the case RSSI>20% were taken into account.

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Sformatowano: Angielski (Wielka Brytania)

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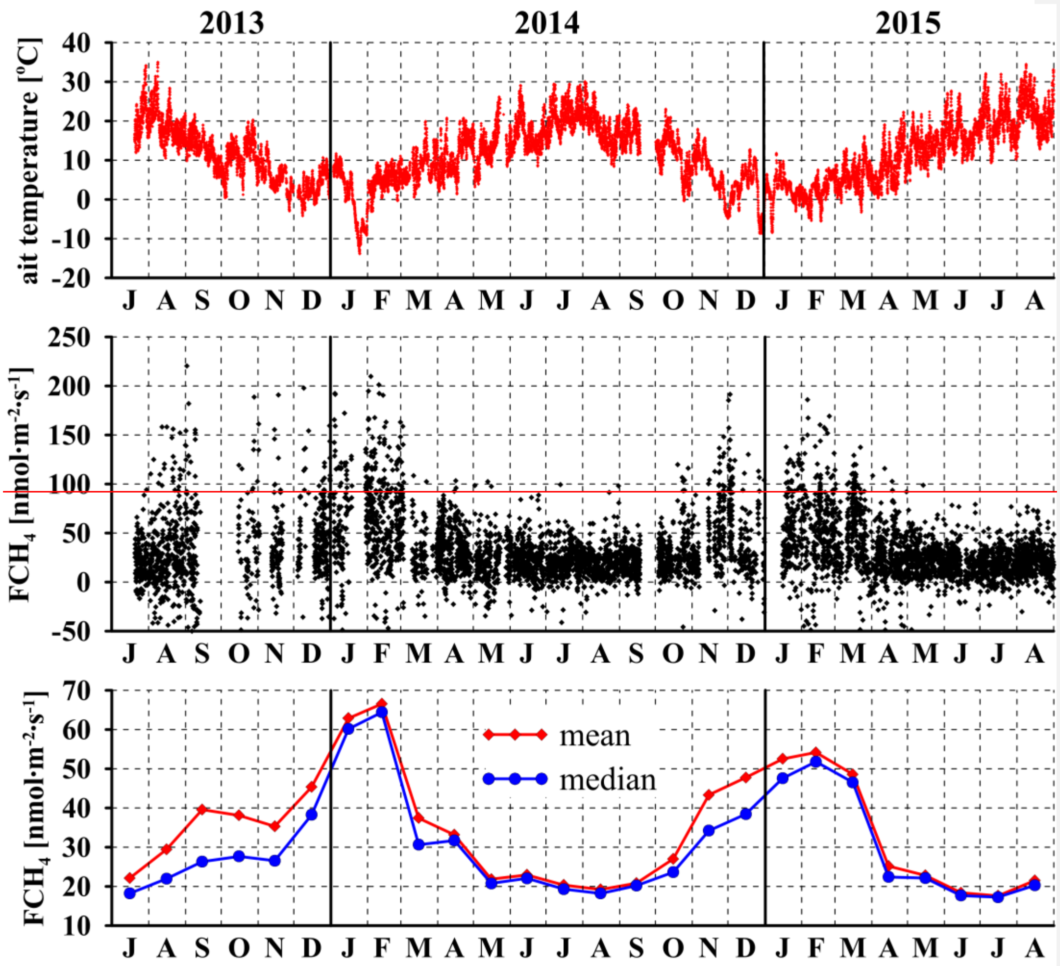
2013
2014
2015
ait temperature [ºC]
40
30
20
10
0
-10
-20
J A S O N D J F M A M J J A S O N D J F M A M J J A
FCH_4 [nmol·m^{-2}·s^{-1}]
250
200
150
100
50
0
-50
J A S O N D J F M A M J J A S O N D J F M A M J J A
FCH_4 [nmol·m^{-2}·s^{-1}]
70
60
50
40
30
20
10
mean
median
J A S O N D J F M A M J J A S O N D J F M A M J J A

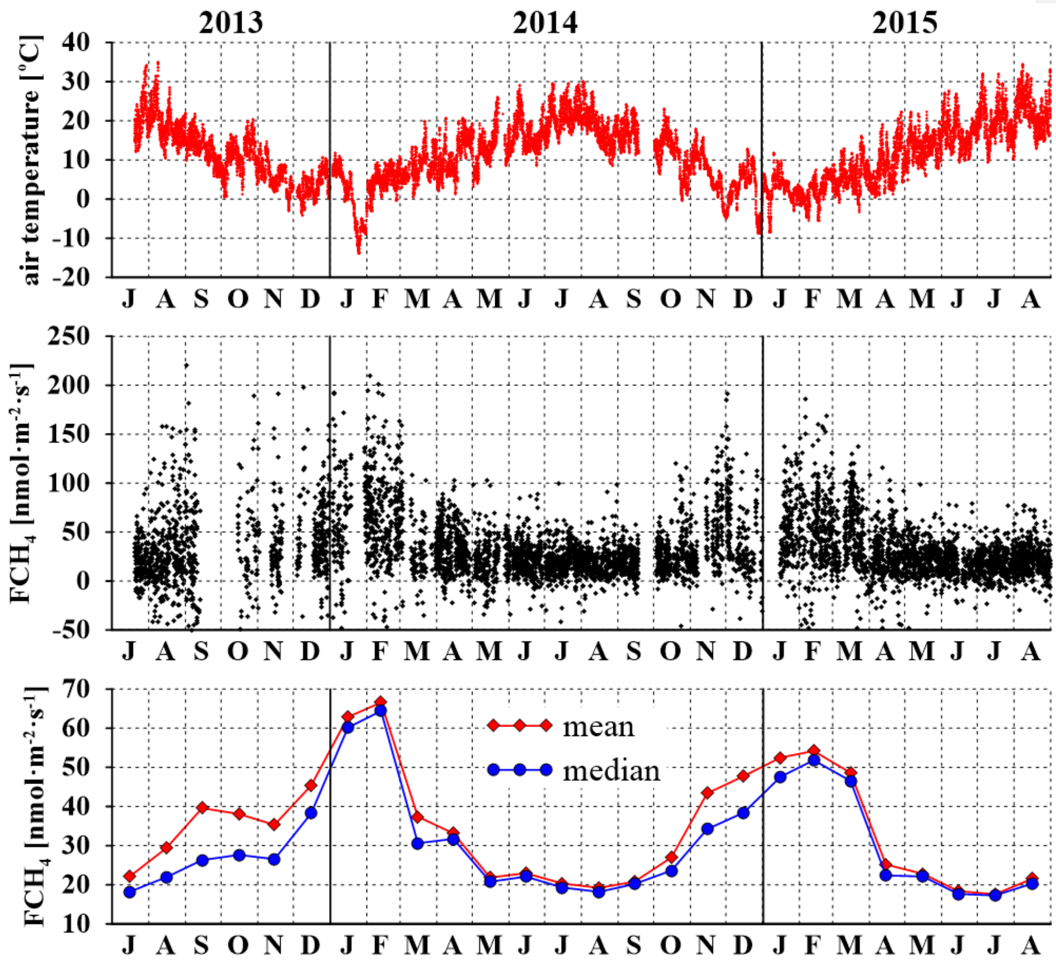

Figure 3. One hour means of air temperature, one hour net turbulent FCH_4 flux (approved by three stationarity tests), mean and median of FCH_4 measured in the ~~Łódź centre in the~~ period July 2013 – August 2015.

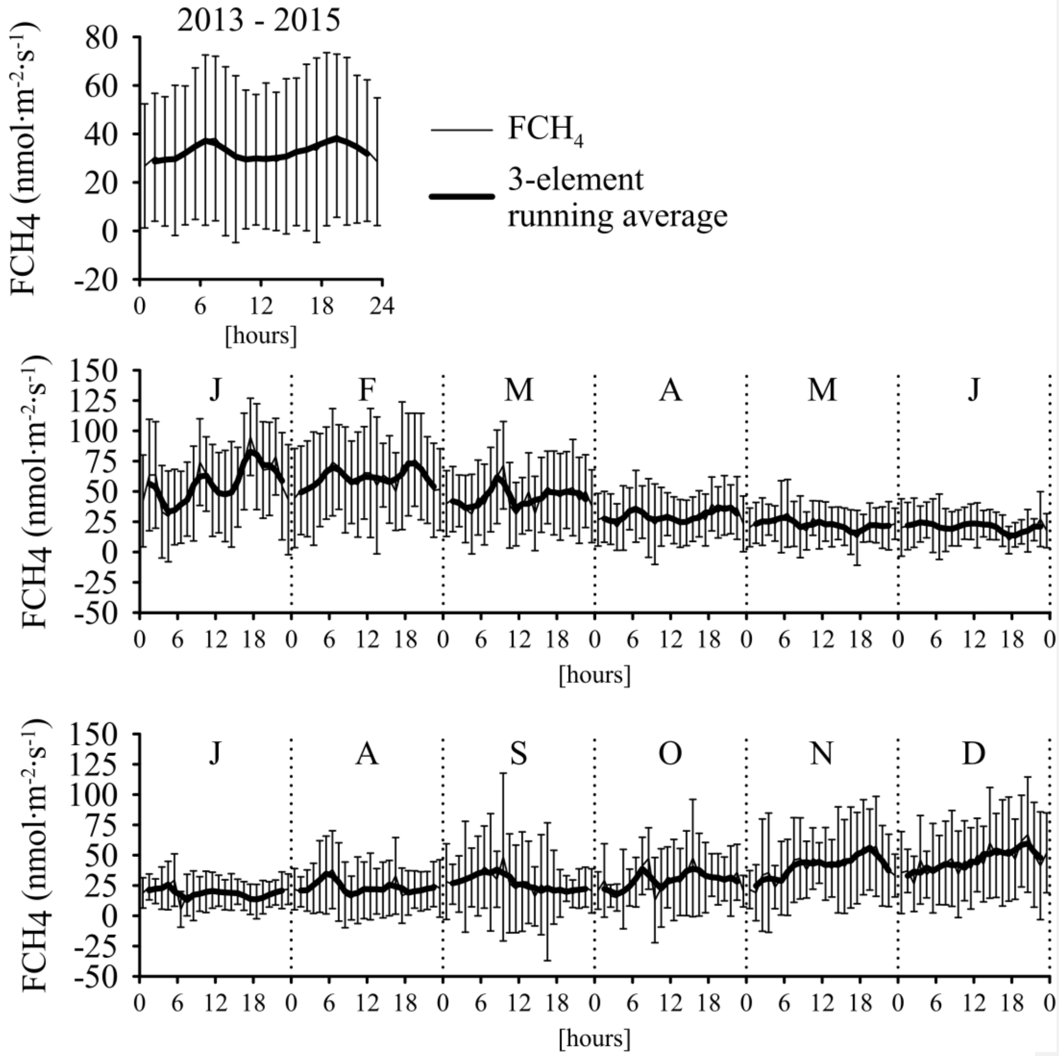

Figure 4. Mean diurnal variability of FCH_4 flux~~ in the Łódź centre~~ in the period July 2013 – August 2015 (top left figure) and for months. Thin and thick black lines indicate, respectively, variability of FCH_4 and 3-element running average of FCH_4. Thin vertical lines indicate standard deviation of FCH_4.

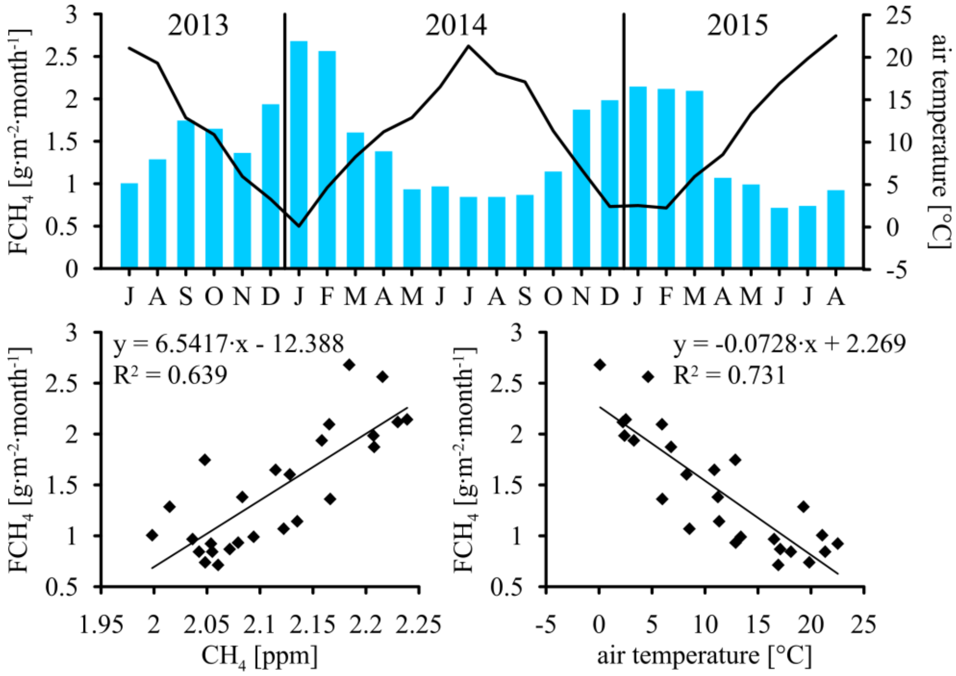

Figure 5. Monthly totals of FCH_4 (top) in relation with mean monthly CH_4 concentration (bottom left) and mean monthly air temperature (bottom right) in the ~~Łódź centre in the~~ period July 2013 – August 2015.

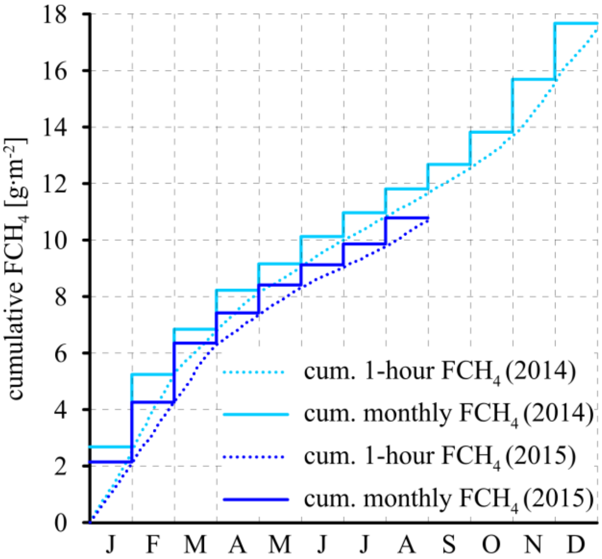

Figure 6. Cumulative fluxes of FCH_4 in the ~~centre of Łódź in the~~ period January – December 2014 (~~blues~~light blue lines) and January – August 2015 (~~navy~~dark blue lines). Dotted and solid lines indicate, respectively, cumulative annual FCH_4 calculated on the basis of all 1-hour data and on the basis of integrated mean daily courses of consecutive months.

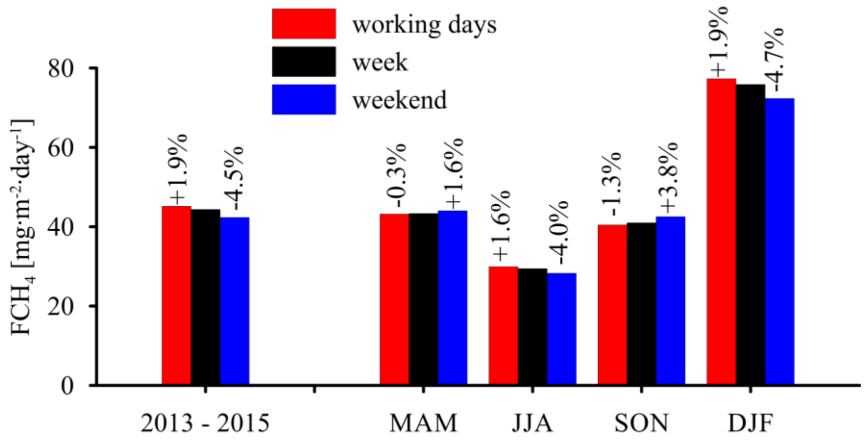

Figure. 7. Mean daily values of FCH_4 ~~in the centre of Łódź~~ in the period July 2013 – June 2015 calculated for study period and seasons. Red, black and blue bars indicate, respectively, mean daily exchange during working days (Monday to Friday), weeks (Monday to Sunday) and weekends (Saturday and Sunday).

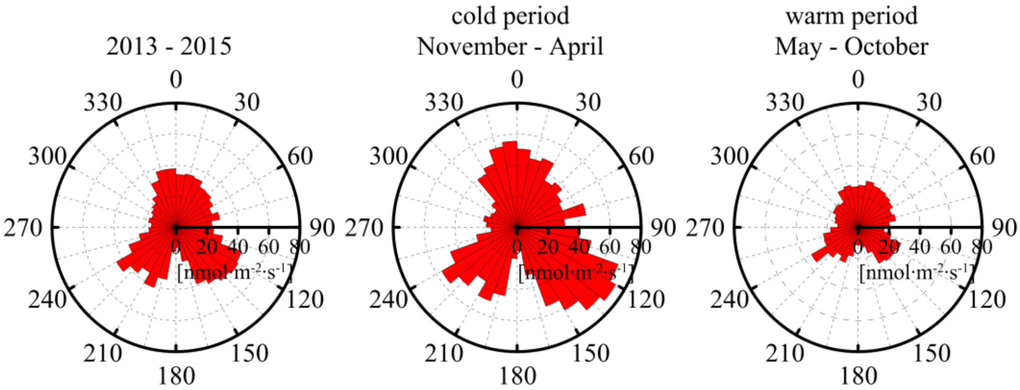

Figure 8. Mean FCH_4 calculated for all data (left), cold (middle) and warm period (right) in relation with wind direction in the ~~centre of Łódź in the~~ period July 2013 – August 2015.

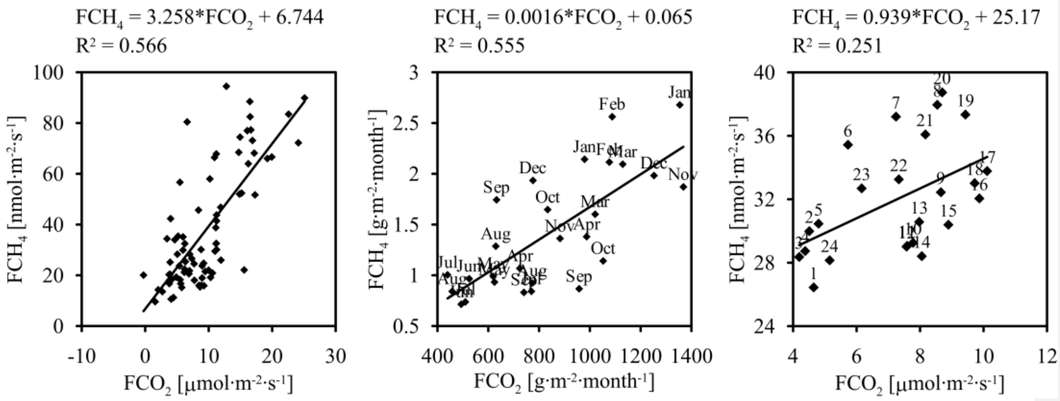

Figure 9. Mean daily (left), monthly (middle) and hourly FCH_4 fluxes against FCO_2 in the ~~Łódź centre~~period July 2013 – June 2015.

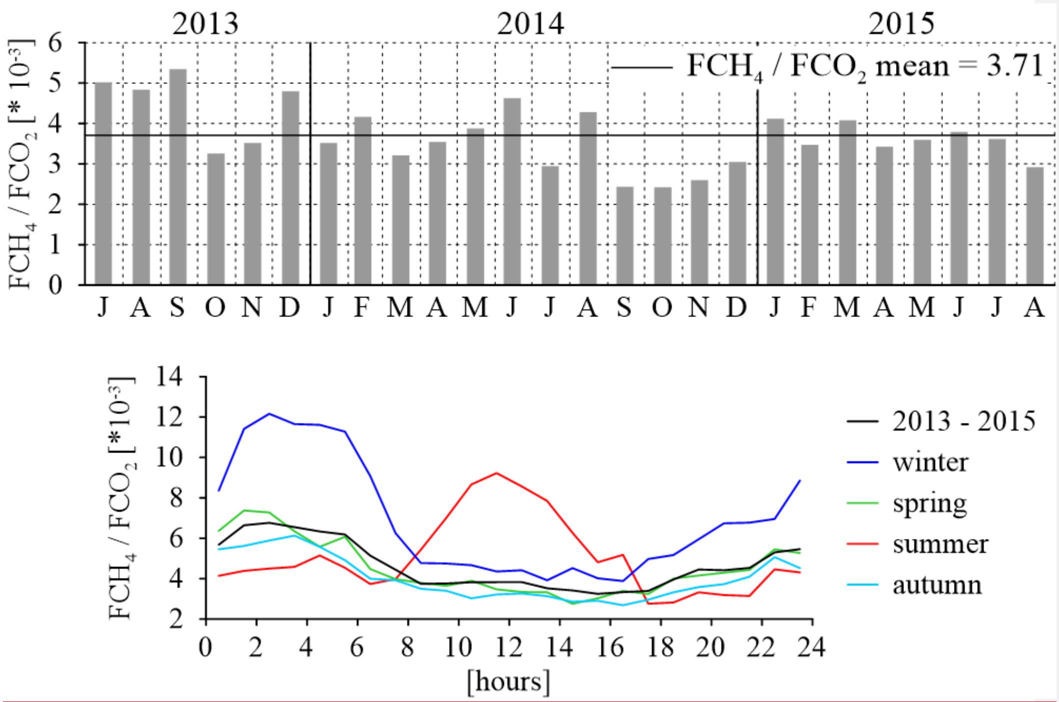

Fig. 10. Monthly FCH_4 to FCO_2 ratio (up) and mean diurnal courses of FCH_4 to FCO_2 ratio in the period July 2013 – ~~June~~August 2015 and for seasons.