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 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. Before uploading the revised manuscript we will make every effort to ensure the quality of language is as high as possible. 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 page 12 line 3 and page 13, line 10 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:

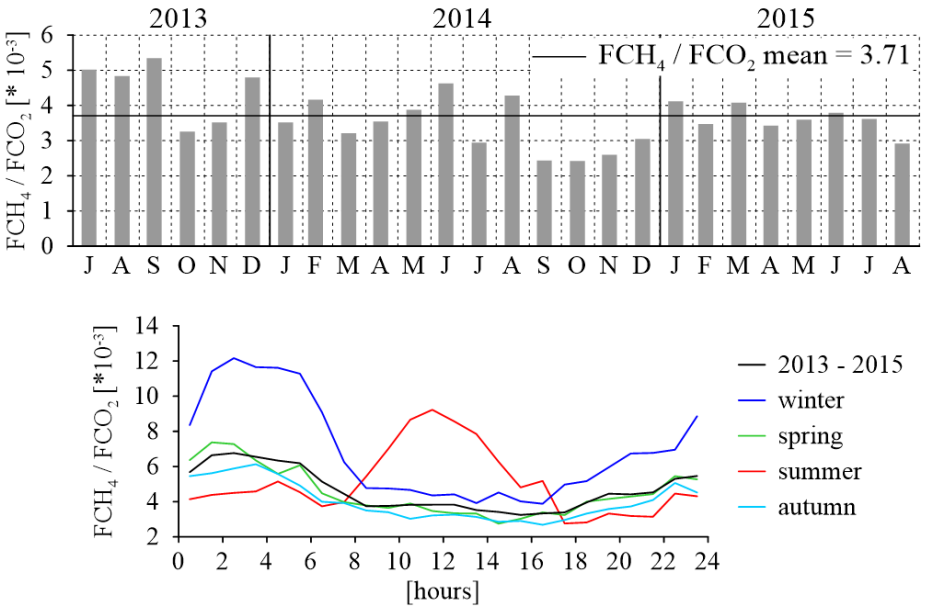

Fig. 10. Monthly FCH4 to FCO2 ratio (up) and mean diurnal courses of FCH$ to FCO2 ratio in the period July 2013 – August 2015 and for seasons.

*"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.

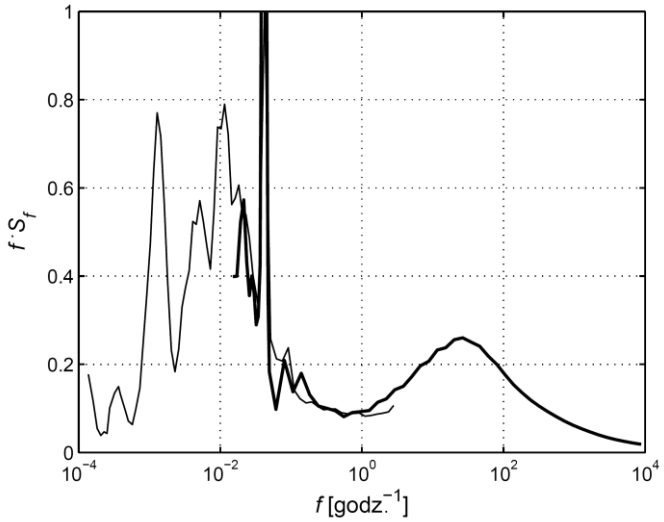

Fig. Wind speed spectrum at Lipowa measurement station in Łódź. Left thin line – on the base of cup anemometer (10 min data from years 2001 -2002), Right bolt line – on the base of sonic anemometer data (10 Hz) from the period 12.06.2002–4.08.2002. ("godz." means "hour" in Polish)

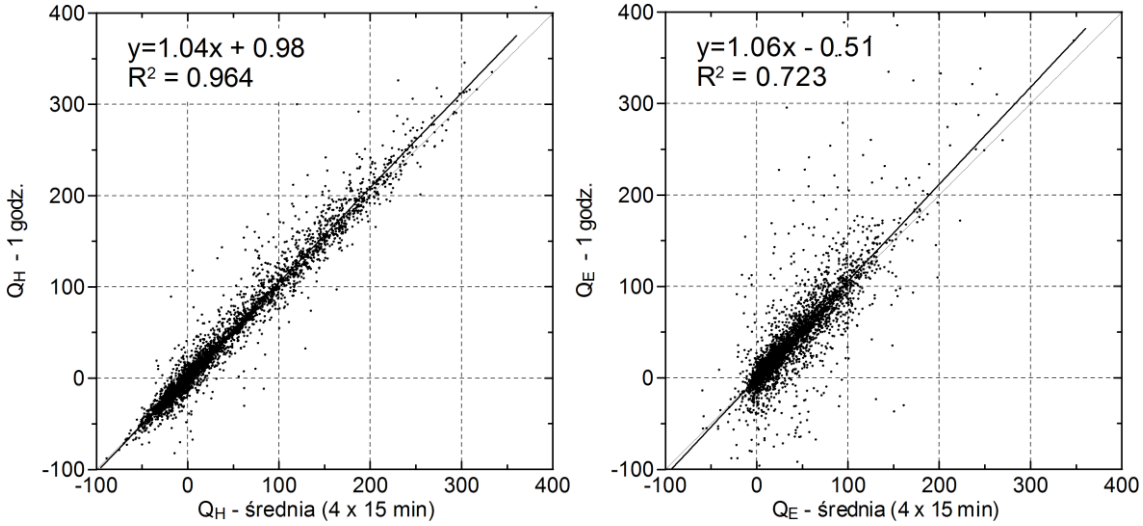

Fig. 2 The comparison of turbulent fluxes of sensible, QH, and latent, QE heat calculated for 1 hour averaging period (Q – 1 godz.) and as a mean of 4 values for 15-min averaging period (Q – średnia (4x15min)). Lipowa measurement station years 2000-2002.

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.