Response to Anonymous Referee #2

We would like to thank the reviewer for helpful comments. Please find our response to the reviewer's comments in blue in the following.

General comments:

1. The approach represents a new application of ground-based open-path remote sensing to estimate GHG emissions from an urban area and will likely be of interest to the atmospheric science community. The paper is reasonably well written though could be substantially improved in terms of both technical completeness and clarity. In particular, the paper suffers from several sections with unclear writing and sections which miss key points regarding the range of assumptions required to derive the results that are reported (see comments below).

   In addition, the paper promotes a future space mission. This seems inappropriate given that the observing strategy from space will yield very dilute optical paths compared to those obtained from the mountaintop. I suggest reducing the emphasis on the satellite (e.g., Section 4.3) or adding additional quantitative information regarding the differences between the observing strategies.

   Response: We have considered the reviewer's comments and edited the paper to address the reviewer's concerns on the unclear sections (please refer to the specific comments below).

   Regarding the satellite emphasis in Section 4.3, we believe that our study is very applicable to future geostationary satellite missions and that this subject is addressed in the appropriate depth in the paper on the similarities and differences between the observing strategies on Mount Wilson and from space. We believe that it is sufficient to introduce this topic in the present paper, leaving detailed discussions about lessons learned from CLARS observations to future papers in preparation focused on radiative transfer and aerosol effects, comparative retrieval precisions for GHGs, and tradeoffs related to spatial and spectral resolution.

2. The paper weakly supports the uncertainty estimates on CH_4 emissions. I suggest the authors consider and address how each of the sources of uncertainty are estimated and justified. First, can CO_2 and CH_4 emissions from the LA Megacity be estimated with stated accuracy from the product of California's total GHG emissions weighted by the fraction of CA's population residing in the MegaCity? Please include this in the assumptions section (4.1) and discuss the following: - what is the definition of the spatial domain being considered at the MegaCity? This affects not only the population being considered but also the relative contributions of CO_2 and CH4 sources. - Why aren't agricultural CH_4 emissions

included if the domain includes Chino, CA. - What is the justification for omitting biosphere CO_2 fluxes in the estimate of CO_2 exchange, particularly in winter ? - what is the justification for suggesting that Mega City CO_2 fluxes are proportional to the fraction of CA population known to within 10% ?

Response: We have edited the paper to address the calculations and uncertainties of the bottom-up CH_4 emissions better. Please see our responses to comments #11 and #12.

We have included the definition of the spatial domain of Los Angeles megacity in our paper as requested. On page 17052 line 15, after the sentence "With the assumptions described in the previous subsection, we estimate the top-down annual CH_4 emission for the Los Angeles megacity based on the CLARS-FTS observations.", we added "In this analysis, we define the Los Angeles megacity as the spatial domain of the South Coast Los Angeles basin."

Regarding the estimation of the bottom-up emissions, because the California Air Resources Board does not provide GHG emissions on district or county level, we need to estimate the emissions for the South Coast Los Angeles basin from the statewide emissions. There are some uncertainties involved when scaling the statewide emission by population. However, we believe that it is appropriate to estimate CO_2 emission in the basin by apportioning the statewide emission using population because fossil fuel combustion is the main source of anthropogenic CO_2. Wunch et al. (2009) used the same method to estimate CO_2 emissions in the South Coast Los Angeles basin and found the estimated CO_2 emissions are consistent with the EDGAR database. Therefore, we believe a 10% uncertainty is reasonable for our estimates.

Regarding the biosphere CO_2 fluxes, the California Air Resources Board bottom-up emission inventory does not include the biosphere sector. In our analysis, we assume that the biosphere has a negligible impact. This is a reasonable assumption since fossil fuel combustion dominates total CO_2 emission (at least 95-99%) in the Los Angeles basin even in winter seasons according to analysis provided by our colleague Meemong Lee at JPL. Her analysis is based on the fossil fuel CO_2 emission from Vulcan and biogenic CO_2 flux from CASA-GFED.

3. Second, how are the XCH4xs/XCO_2xs slope estimated? Does this assume all errors are random among the 27 paths (6.4±0.5 ppb CH_4 (ppm CO_2)$^{-1}$) to within ~8%. This is not discussed in the text or justified in any manner. In particular, the uncertainty Figure 5 shows regions with higher (e.g., Montebello, Walnut, Yorba Linda, Fullerton) and lower (Hollywood, East Los Angles, Long Beach, Palo Verdes) XCH_4xs/XCO_2xs slopes. This doesn't support the implicit assumption of random error in the variation of XCH4xs/XCO_2xs slopes. It would seem more appropriate to state an upper estimate of systematic uncertainties that includes the range of slopes obtained across sites. Also, the assumption of negligible bias in

XCH_4xs/XCO_2xs slope due to aerosols is needs at least some simple quantitative justification.

Response: The $XCH_{4(XS)}/XCO_{2(XS)}$ slopes for each reflection points were obtained using the orthogonal distance regression (ODR) analysis. ODR analysis takes into account of both uncertainties in the y and x variables. To clarify this better, on page 17047 line 23, we expanded the sentence from "We used orthogonal distance regression (ODR) analysis of $XCH_{4(XS)}/XCO_{2(XS))}$ to quantify the emissions of CH_4 relative to CO_2 in the Los Angeles megacity." to "We used orthogonal distance regression (ODR) analysis, which considers uncertainties in both $XCH_{4(XS)}$ and $XCO_{2(XS)}$, to quantify the emissions of CH_4 relative to CO_2 in the Los Angeles megacity."

We clarified in the text that the uncertainty for the average $XCH_{4(XS)}/XCO_{2(XS)}$ ratio in the basin is the standard deviation. Please refer to our response to comment #12.

The quantitative justification of the assumption of negligible bias in $XCH_{4(XS)}/XCO_{2(XS)}$ slope due to aerosol has been added to the text. Please read our response to comment #9.

4. Last, please expand observations and emissions estimates sections to include description of the in-situ measurements at Mt Wilson and Pasadena that are included in Table 4.

Response: We have added explanation of in situ measurements at Mt. Wilson and Pasadena in the text as requested.

On page 17049 line 8, we added the following text "At California Institute of Technology (Caltech) in Pasadena and at the CLARS facility on Mount Wilson, in situ CH_4 and CO_2 mixing ratios were measured by two Picarro G1301 CO_2-CH_4 analyzers (Newman et al., 2013). Secondary standards, calibrated against primary NOAA standards, were run every 11 hours. Because of the complex boundary layer dynamics near mountains, measurements on Mount Wilson is influenced by upslope flow of air mass from the basin during the day while expose to the clean background air from the free tropospheric at night (Hsu et al., 2009). Using the mean of hourly averages from 22:00 – 03:00 PST on Mount Wilson as the background reference, CH_4 and CO_2 excess mixing ratios were calculated by subtracting the background reference from the daytime hourly averaged measurements at Mount Wilson and at Caltech. The ratios were the correlation slopes between the two."

On page 17052 line 12, we modified the sentence from "This is in good agreement with recent studies (Wunch et al., 2009; Hsu et al., 2010; Wennberg et al., 2012; Peischl et al., 2013; Jeong et al., 2013)." to "This is in good agreement with the top-down CH_4 emissions from recent studies (Wunch et al., 2009; Hsu et al., 2010; Wennberg et al., 2012; Peischl et al., 2013; Jeong et al., 2013) and the CH_4 emissions derived from the observations at Caltech and on Mount Wilson (using the same bottom-up CO_2 emissions for the Los Angeles basin)."

Specific comments:

5. Abstract. Where does the uncertainty in inventory-based CH_4 emissions derived?

   Response: We do not quite understand this question because we did not report the inventory-based CH_4 emission in the abstract. We reported the CH_4:CO_2 ratio in Los Angeles based on the California Air Resource Board bottom-up emission inventory to be 4.6 ± 0.9 ppb CH_4 (ppm CO_2)$^{-1}$ in the abstract. The uncertainties are calculated assuming a 10% uncertainty in the statewide total CH_4 and CO_2 emissions and another 10% uncertainty in apportioning the statewide emission to Los Angeles basin emission by population. In addition, we also mentioned in the abstract that the derived top-down CH_4 emission based on our Mount Wilson FTS observations is 0.39 ± 0.06 Tg CH_4 year^{-1}. The uncertainties are derived based on the uncertainties in the bottom-up CO_2 emission in Los Angeles and the $XCH_{4(XS)}/XCO_{2(XS)}$ ratio observed by CLARS-FTS.

   No changes have been made in the text for this comment.

6. pg. 17040, line 15. Please qualify the statement to include the expected accuracy obtained using 8 point observing sites.

   Response: We have made changes in the statement as requested. The statement is changed from "Kort et al. (2013) concluded that the size and complexity of the Los Angeles megacity urban dome requires a network of at least eight strategically located continuous surface in situ observing sites to quantify and track GHG emissions over time." to "Kort et al. (2013) concluded that the size and complexity of the Los Angeles megacity urban dome requires a network of at least eight strategically located continuous surface in situ observing sites to quantify and track GHG emissions over time with ~10% uncertainty."

7. pg 17049, line 1. likely typo: "are DUE to ..."

   Response: Thank you for catching this. We have corrected this in the text.

8. Section 4.1 Assumptions

Assumptions 1&2. While likely true, the reasons for including assumptions 1&2 are not clearly motivated. Please add statements for each, clearly identifying why it matters to the emissions analysis.

Response: The motivation for assumption 1&2 is mentioned in Section 3.2 of the paper "Several studies have reported strong correlations between CH_4 and CO_2 measured in the PBL in source regions (Peischl et al., 2013; Wennberg et al., 2012; Wunch et al., 2009; S. Newman, personal communication, 2014). Slopes of CH_4:CO_2 correlation plots have been identified with local emission ratios for the two gases. Since the uncertainties in CH_4 emissions are considerably larger than that in CO_2 emissions, we may use the correlation slope to reduce the CH_4 emission uncertainties."

We agree with the reviewer that it may not appear very clear about the motivations for assumption 1 and 2. Therefore, following the above paragraph in Section 3.2, we added the sentence "A few assumptions are used when quantifying CH_4 emission based on CH_4:CO_2 correlation. These assumptions will be discussed in Section 4.1 of the paper."

9. Assumption 3. Are aerosol biases in the background subtracted column ratios $XCH_{4(XS)}$:$X_{CO2(XS)}$ small enough to not compromise analysis for emissions ? The paper must include a quantitative estimates or at least an upper limit on this bias.

   Response: We have considered the reviewer's comment and performed a quantitative estimates for the aerosol impact on XCH_4:XCO_2. Zhang et al. (2014) expanded their aerosol analysis on XCH_4 and XCH_4:XCO_2 and found that aerosol impact on XCH_4 and XCO_2 is nearly completely canceled out in XCH_4:XCO_2. In our study, aerosol impact in XCH_4:XCO_2 ratio is expected to be <0.5%. On page 17050 line 17, we edited the following sentence "Since the CO_2 and CH_4 observations used in this analysis are retrieved at nearly identical wavelengths (1.61μm vs. 1.66μm), the aerosol-induced bias on XCO_2 and XCH_4 should be nearly identical and canceled out in the ratio." to "Further analysis based on Zhang et al. (2014) indicates that the aerosol-induced bias on XCO_2 and XCH_4 is nearly identical and cancel out in the ratio since the CO_2 and CH_4 observations used in this analysis are retrieved at nearly identical wavelengths (1.61μm vs. 1.66μm). The uncertainty of XCH_4:XCO_2 ratio due to aerosol is negligible (<0.5%)."

10. Assumption 4. How much data is retained after filtering in each season? How are uncertainties propagated into annual mean ?

    Response: The fraction of data passing through the data filter varies by a factor of two in different seasons. We believe that the seasonal bias in our analysis is small. This seems to be a reasonable assumption since tight correlation is observed between $XCH_{4(XS)}$ and $XCO_{2(XS)}$ throughout the year, the contribution of seasonal sampling bias, if any, has a negligible effect on the random error of the annual average $XCH_{4(XS)}$:$XCO_{2(XS)}$ correlation slope.

We have revised assumption 4 to "Seasonal bias in the $XCH_{4(XS)}$:$XCO_{2(XS)}$ correlation slope is small. Certain times of the year are more likely to be influenced by cloud and aerosol events in Los Angeles and have correspondingly fewer measurements that pass the data quality filters. The fraction of data passing through the data filter varies by a factor of two in different seasons. In our analysis the effect of seasonal bias is small. This seems to be a reasonable assumption since tight correlation was observed between $XCH_{4(XS)}$ and $XCO_{2(XS)}$ throughout the year, the contribution of seasonal sampling bias, if any, has a negligible effect on the random error of the annual average $XCH_{4(XS)}$:$XCO_{2(XS)}$ correlation slope."

11. pg 17051, line 27. The bottom-up estimate of CH_4 emissions is unclear. Why are agricultural CH_4 emissions subtracted from CARB inventory. There are non-zero CH_4 emissions expected from dairies in the Chino area.

    Response: We estimated that agriculture contributes only a small portion of CH_4 emissions. According to the California Air Resources Board, agriculture and forestry contributes to 62% of total methane emission in the state. The Los Angeles basin contains less than 2% of farmlands in California according to the United States Department of Agriculture. Therefore, methane emissions came from agriculture and forestry in Los Angeles basin only contributes to ~1% of the total statewide CH_4 emissions. This method has also been used in Wunch et al. (2009) and Peischl et al. (2013). We revised the explanation of the bottom-up estimate of CH_4 emission in the text. On page 17051, line 27, we revised the following text "For the bottom-up CH_4 emission in the Los Angeles megacity, we used the same method as in Wunch et al. (2009) and Peischl et al. (2013). That is, subtracting agriculture and forestry sector from the total statewide emission, then apportioned by population." to "For the bottom-up CH_4 emission in the Los Angeles megacity, we used the same method as in Wunch et al. (2009) and Peischl et al. (2013). Agriculture and forestry contributes 62% of total CH_4 emission in the state (California Air Resources Board, 2011) but the Los Angeles basin contains less than 2% of farmlands in California (United States Department of Agriculture, 2011). Therefore we estimated the bottom-up CH_4 emissions in the basin by subtracting agriculture and forestry sector from the total statewide emission then apportioned by population."

12. pg 17052, line 9. How is 0.06 Tg CH_4 yr^{-1} uncertainty CH_4 emissions obtained ? Uncertainties in bottom-up CO_2 emissions was estimated as 166±23 Tg CO_2 year – 1 (more like sqrt(2) * 10%~14%). Also, as above, how was uncertainty in XCH_{4xs}/XCO_{2xs} slope obtained?

Response: The uncertainty of our top-down CH_4 emission is derived using error propagation of the uncertainty of the bottom-up CO_2 emission (±23 Tg CO_2/year) and the uncertainty of the average XCH_4(xs)/XCO_2(xs) slope in the Los Angeles basin. Standard deviation of the observed CLARS-FTS XCH_4(xs)/XCO_2(xs) slopes among the 28 reflection points is used as the uncertainty of the average XCH_4(xs)/XCO_2(xs) slope in the Los Angeles basin.

We have made the following changes in the text to explain our calculations in a better way:

- We clarified in the text that the uncertainty for the average $XCH_{4(XS)}/XCO_{2(XS)}$ ratio in the basin is the standard deviation. On page 17048 line 9, we modified the sentence "The mean for all 28 reflection points was 6.4±0.5 ppb CH_4 (ppm CO_2)$^{-1}$ with individual values ranging from 5.4 to 7.3 ppb CH_4 (ppmCO_2)$^{-1}$." to "The mean ± one standard deviation for all 28 reflection points was 6.4±0.5 ppb CH_4 (ppm CO_2)$^{-1}$ with individual values ranging from 5.4 to 7.3 ppb CH_4 (ppm CO_2)$^{-1}$."
- We included the values and uncertainties when explaining our calculation of top-down CH_4 emission. On page 17052 line 2, we modified the sentence "Using the bottom-up emission inventory of CO_2 for the Los Angeles megacity and the CH_4:CO_2 ratio observed by the CLARS-FTS, we derived the CH_4 emission inventory using Eq. (3), where $E_{CH4 \mid top\text{-}down}$ is the top-down CH_4 emissions inferred by the CLARS-FTS observations, $E_{CO2 \mid bottom\text{-}up}$ is the bottom-up CO_2 emissions, $XCH_4/XCO_{2 \mid slope}$ is the $XCH_{4(XS)}/XCO_{2(XS)}$ ratio observed by the FTS and M_{CH4}/MW_{CO2} is the ratio of molecular weight of CO_2 and CH_4." to "Using the bottom-up emission inventory of CO_2 for the Los Angeles megacity (166±23 Tg CO_2 year^{-1}) and the average CH_4:CO_2 ratio observed by the CLARS-FTS (6.4±0.5 ppb CH_4 (ppmCO_2)$^{-1}$), we derived the CH_4 emission inventory using Eq. (3), where $E_{CH4 \mid top\text{-}down}$ is the top-down CH_4 emissions inferred by the CLARS-FTS observations, $E_{CO2 \mid bottom\text{-}up}$ is the bottom-up CO_2 emissions, $XCH_4/XCO_{2 \mid slope}$ is the average $XCH_{4(XS)}/XCO_{2(XS)}$ ratio observed by the FTS and MW_{CH4}/MW_{CO2} is the ratio of molecular weight of CO_2 and CH_4 (that is, 16 g CH_4/ 44 g CO_2)."

13. pg 17052, lines 14-20. The statements concerning spatial variation in XCH_{4xs}/XCO_{2xs} slopes suggests uncertainties are likely greater than estimated from Eq (3). It would appear more appropriate to state a range of CH_4 emissions assuming the range of slopes obtained.

Response: We think the reviewer might have misunderstood these statements. The statements indicate that due to the spatial variation in XCH_4(xs)/XCO_2(xs) slope across the basin, if we use observations from only one location, it can lead to a bias in the derived emissions for the entire basin. Therefore, it is important to have a robust measurement technique like CLARS-FTS which provides spatiotemporal coverage of the basin over time to have a more appropriate quantification for the entire basin.

To quantify emissions for the basin, we used the average of the XCH_4(xs)/XCO_2(xs) slopes observed for the 28 reflection points and defined the uncertainty of the average XCH_4(xs)/XCO_2(xs) slope for the Los Angeles basin as the standard deviation among the XCH_4(xs)/XCO_2(xs) slopes observed for the 28 reflection points instead of the range. We believe that standard deviation represents the uncertainty of the slope.

No changes have been made in the text for this comment.

14. Table 4. Why are there two CH_4 emissions results (0.40±0.10 and 0.60±0.10) reported for Wunch et al? In addition, the previous study by Hsu et al. (2009) used methane and carbon monoxide (not carbon dioxide) measurements to compute CH_4:CO slopes and CH_4 emissions. Is new data being reported from the work of Hsu et al (2009) and here in Table 4?

    Response: There are two CH_4 emission results reported by Wunch et al. (2009). 0.40±0.10 Tg CH_4/ year is the top-down CH_4 emission estimated based on their CH_4:CO ratios while 0.60±0.10 Tg CH_4/ year is the emission estimated based on their CH_4:CO_2 ratios. We have clarified this in the caption of Table 4 by adding the following sentence "Wunch et al. (2009) reported two top-down CH_4 estimates: 0.40±0.10 Tg CH_4/ year derived from CH_4:CO_2 ratio and 0.60±0.10 Tg CH_4/ year derived from CH_4:CO ratio."

    The CH_4:CO_2 ratio from Mount Wilson was calculated based on a more recent data set on Mount Wilson. To clarify this, we have added explanation of in situ measurements at Mt. Wilson in the text. Please refer to our response to comment #4.

15. Figure 5. Please mark the location of Mt Wilson on maps.

    Response: Location of Mount Wilson is added on maps in Figure 5 as requested.