

Response to Anonymous Referee #1

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. My one general question deals with the uncertainty of comparing the reflected beam vs. the direct beam. How would a residual boundary layer below the altitude of Mt. Wilson affect the uncertainty of the excess CH₄ or CO₂ abundance? Along those lines, how would spatial variability in CH₄ or CO₂ in the atmosphere above 1.6 km affect the excess ratio?

Response: We define the excess CH₄ and CO₂ mixing ratios as the excess due to the column below Mount Wilson, instead of the excess in the boundary layer. Therefore, any residual boundary layer below Mount Wilson will not be subtracted when calculating CH₄ and CO₂ excess. We do not expect this to impact the CH₄:CO₂ excess ratio in the Los Angeles basin. Because a residual boundary layer consists of air mass from the previous daytime boundary layer, the relative CH₄:CO₂ ratio in the air mass would still carry information about the emission ratio of CH₄:CO₂ from the basin in the previous day. In addition, the residual layer tends to mix into the boundary layer as the boundary layer grows during the day. We do not expect this to impact our CH₄:CO₂ excess ratio for a two-year period.

Spatial variation in CO₂ and CH₄ mixing ratio above Mount Wilson in the basin is possible due to entrainment of boundary layer air mass into the free troposphere and long-range transport. However, this is expected to have a negligible impact in the excess ratio. We have added this assumption in the text as requested. Please see our response to comment #4 below.

Specific comments:

2. p. 17039, Line 15; do you have a citation for the $\pm 10\%$ uncertainty?

Response: Yes, we have included the citation, de la Rue du Can et al. (2008). The following is added to the list of references "de la Rue du Can, S., Wenzel, T., and Price, L.: Improving the Carbon Dioxide Emission Estimates from the Combustion of Fossil Fuels in California, Report prepared for the California Air Resources Board and the California Environmental Protection Agency, 2008." Changes can be found on page 3 line 1 and page 17 line 14 of the revised manuscript.

3. p. 17040, Line 2; You should clarify that a column measurement is less influenced by local sources as long as these sources' emissions don't fill the boundary layer.

Response: We have given some thought about this comment. However, it seems very unlikely that there are any CO₂ or CH₄ source, which fill the boundary layer. We clarified this by editing the sentence on page 17039 line 28 (page 3 line 14 of the revised manuscript) from "Since column measurements are relatively insensitive to boundary layer height variations and are less influenced by local sources than ground in situ measurements, they should be more representative of the area." to "Since column measurements are relatively insensitive to boundary layer height variations and are less influenced by local surface sources, than ground in situ measurements, they should be more representative of the area."

4. p. 17041, Section 4.1; Please add some discussion on the authors' assumptions for the slant column density of a gas above Mt. Wilson.

Response: We included an additional bullet point in Section 4.1. On page 17051 line 13 (page 13 line 19 of the revised manuscript), the following text has been included "Spatial variation in the atmospheric column of CO₂ and CH₄ above Mount Wilson is minimal and does not affect the XCH₄:XCO₂ excess ratio. Spatial variation in CO₂ and CH₄ mixing ratio above Mount Wilson in the basin is possible due to entrainment of boundary layer air mass into the free troposphere and long-range transport. It can be shown that spatial variability in the column above Mount Wilson due to entrainment of boundary layer height or long-range transport adds less than 1% uncertainty to XCH₄:XCO₂ excess ratio."

5. Table 3; I don't see where this table is cited in the text.

Response: We cited Table 3 in the text by adding the following sentence on page 17048 line 7 (page 10 line 13 of the revised manuscript) "Table 3 lists the correlation slopes and their uncertainties for the 28 basin reflection points."

6. Table 3; The uncertainties in this table should include the accuracy uncertainties as well as the fit uncertainties. This overall uncertainty should then be used in the average for the entire basin.

Response: We believe that it is sufficient to include the accuracy uncertainties in the text and as footnote in Table 3. In table 3, we added a footnote "The uncertainties include only fitting uncertainties. Systematic uncertainties of ~4% were not taken account here (Fu et al., 2014)." This change can be found on page 24 of the revised manuscript.

7. Table 4; Why not add the 2010 CalNex CH₄:CO₂ from Wennberg et al. (2012)?

Response: We have added the 2010 CalNex CH₄:CO₂ ratio (0.66 ± 0.03 ppb CH₄/ppb CO₂) and the reported CH₄ emission (0.44 ± 0.1 Tg CH₄/ year) from Wennberg et al. (2012) in Table 4. These changes can be found on page 25 of the revised manuscript.

8. Technical corrections “in-situ” is sometimes hyphenated, sometimes not throughout the paper.

Response: Thank you for pointing this out. “In-situ” has been changed to “in situ” throughout the paper.

9. p. 17049, line 7; you seem to have one too many citations for “Y.-K. Hsu, personal communication”.

Response: We have shortened the citations as requested. The sentence has been edited from “These observations reported ratios ranging from 6.10 to 6.74 ppb CH₄ (ppm CO₂)⁻¹ (Wennberg et al., 2012; Peischl et al., 2013; S. Newman and Y.-K. Hsu, personal communication, 2014; Y.-K. Hsu, personal communication, 2014).” to “These observations reported ratios ranging from 6.10 to 6.74 ppb CH₄ (ppm CO₂)⁻¹ (Wennberg et al., 2012; Peischl et al., 2013; S. Newman, personal communication, 2014; Y.-K. Hsu, personal communication, 2014).” These changes can be found on page 11 line 11 of the revised manuscript.

10. p. 17053, line 19; change text to “an interesting”.

Response: Correction has been made in the text as requested. Please refer to page 16 line 1 of the revised manuscript.

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 CH₄ emissions. I suggest the authors consider and address how each of the sources of uncertainty are estimated and justified. First, can CO₂ and CH₄ 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 CO₂ and CH₄ sources. - Why aren't agricultural CH₄ emissions

included if the domain includes Chino, CA. - What is the justification for omitting biosphere CO₂ fluxes in the estimate of CO₂ exchange, particularly in winter ? - what is the justification for suggesting that Mega City CO₂ 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 CH₄ 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 (page 13 line 27 of the revised manuscript), after the sentence “With the assumptions described in the previous subsection, we estimate the top-down annual CH₄ 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 CO₂ emission in the basin by apportioning the statewide emission using population because fossil fuel combustion is the main source of anthropogenic CO₂. Wunch et al. (2009) used the same method to estimate CO₂ emissions in the South Coast Los Angeles basin and found the estimated CO₂ emissions are consistent with the EDGAR database. Therefore, we believe a 10% uncertainty is reasonable for our estimates.

Regarding the biosphere CO₂ 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 CO₂ 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 CO₂ emission from Vulcan and biogenic CO₂ flux from CASA-GFED.

3. Second, how are the XCH₄_{xs}/XCO₂_{xs} slope estimated? Does this assume all errors are random among the 27 paths (6.4 ± 0.5 ppb CH₄ (ppm CO₂)⁻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 Angeles, Long Beach, Palo Verdes) XCH₄_{xs}/XCO₂_{xs} slopes. This doesn't support the implicit assumption of random error in the variation of XCH₄_{xs}/XCO₂_{xs} 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 $XCH_{4(XS)}/XCO_{2(XS)}$ 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 (page 10 line 4 of the revised manuscript), 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 (page 11 line 12 of the revised manuscript), 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 (page 14 line 24 of the revised manuscript), 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 CH₄ 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 CH₄ emissions derived from the observations at Caltech and on Mount Wilson (using the same bottom-up CO₂ emissions for the Los Angeles basin).”

Specific comments:

5. Abstract. Where does the uncertainty in inventory-based CH₄ emissions derived?

Response: We do not quite understand this question because we did not report the inventory-based CH₄ emission in the abstract. We reported the CH₄:CO₂ ratio in Los Angeles based on the California Air Resource Board bottom-up emission inventory to be 4.6 ± 0.9 ppb CH₄ (ppm CO₂)⁻¹ in the abstract. The uncertainties are calculated assuming a 10% uncertainty in the statewide total CH₄ and CO₂ 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 CH₄ emission based on our Mount Wilson FTS observations is 0.39 ± 0.06 Tg CH₄ year⁻¹. The uncertainties are derived based on the uncertainties in the bottom-up CO₂ 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.” Changes of the revised manuscript can be found on page 3 line 29.

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

Response: Thank you for catching this. We have corrected this in the text. Please refer to page 11 line 6 of the revised manuscript.

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 CH₄ and CO₂ 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 CH₄:CO₂ correlation plots have been identified with local emission ratios for the two gases. Since the uncertainties in CH₄ emissions are considerably larger than that in CO₂ emissions, we may use the correlation slope to reduce the CH₄ 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 CH₄ emission based on CH₄:CO₂ correlation. These assumptions will be discussed in Section 4.1 of the paper.” These changes can be found on page 9 line 19 of the revised manuscript.

9. Assumption 3. Are aerosol biases in the background subtracted column ratios $X_{CH_4(XS)}:X_{CO_2(XS)}$ 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 $X_{CH_4}:X_{CO_2}$. Zhang et al. (2014) expanded their aerosol analysis on X_{CH_4} and $X_{CH_4}:X_{CO_2}$ and found that aerosol impact on X_{CH_4} and X_{CO_2} is nearly completely canceled out in $X_{CH_4}:X_{CO_2}$ (see figure below). In our study, aerosol impact in $X_{CH_4}:X_{CO_2}$ ratio is expected to be <0.5%. On page 17050 line 17 (page 12 line 26 of the revised manuscript), we edited the following sentence “Since the CO₂ and CH₄ observations used in this analysis are retrieved at nearly identical wavelengths (1.61μm vs. 1.66μm), the aerosol-induced bias on X_{CO_2} and X_{CH_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 X_{CO_2} and X_{CH_4} is nearly identical and cancel out in the ratio since the CO₂ and CH₄ observations used in this analysis are retrieved at nearly identical wavelengths (1.61μm vs. 1.66μm). The uncertainty of $X_{CH_4}:X_{CO_2}$ ratio due to aerosol is negligible (<0.5%).”

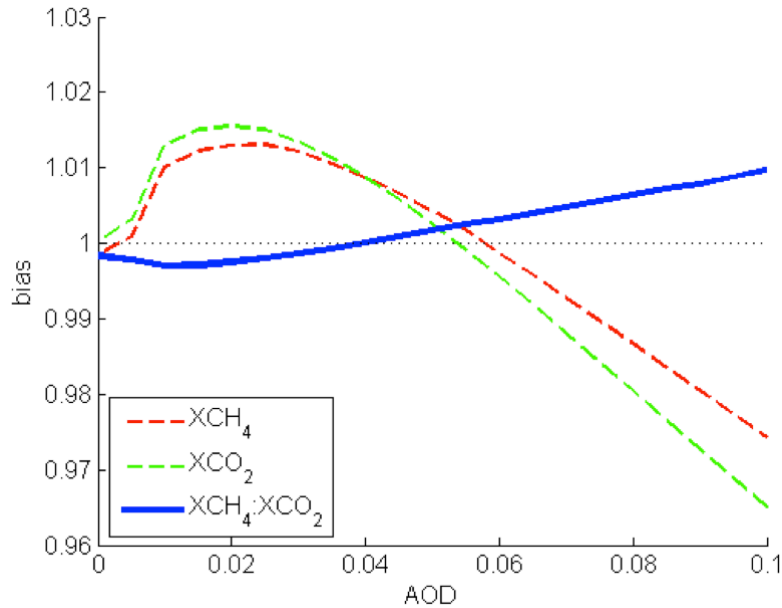


Figure above shows the aerosol bias in XCH₄, XCO₂ and XCH₄:XCO₂ ratio as a function of the aerosol optical depth (AOD) calculated using a radiative transfer model (VLIDORT) by Zhang et al. In our analysis for this paper, AOD is approximately less than 0.01.

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.” These changes can be found on page 13 line 11 of the revised manuscript.

11. pg 17051, line 27. The bottom-up estimate of CH₄ emissions is unclear. Why are agricultural CH₄ emissions subtracted from CARB inventory. There are non-zero CH₄ emissions expected from dairies in the Chino area.

Response: We estimated that agriculture contributes only a small portion of CH₄ 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 CH₄ 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 CH₄ emission in the text. On page 17051, line 27 (page 14 line 8 of the revised manuscript), we revised the following text “For the bottom-up CH₄ 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 CH₄ 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 CH₄ 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, 2012). Therefore we estimated the bottom-up CH₄ 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 CH₄ yr⁻¹ uncertainty CH₄ emissions obtained ? Uncertainties in bottom-up CO₂ emissions was estimated as 166±23 Tg CO₂ year⁻¹ (more like sqrt(2) * 10%~14%). Also, as above, how was uncertainty in XCH_{4(xS)}/XCO_{2(xS)} slope obtained?

Response: The uncertainty of our top-down CH₄ emission is derived using error propagation of the uncertainty of the bottom-up CO₂ emission (±23 Tg CO₂/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 (page 10 line 16 of the revised manuscript), we modified the sentence “The mean for all 28 reflection points was 6.4±0.5 ppb CH₄ (ppm CO₂)⁻¹ with individual values ranging from 5.4 to 7.3 ppb CH₄ (ppm CO₂)⁻¹”

¹.” to “The mean \pm one standard deviation for all 28 reflection points was 6.4 ± 0.5 ppb CH₄ (ppm CO₂)⁻¹ with individual values ranging from 5.4 to 7.3 ppb CH₄ (ppm CO₂)⁻¹.”

- We included the values and uncertainties when explaining our calculation of top-down CH₄ emission. On page 17052 line 2 (page 14 line 15 of the revised manuscript), we modified the sentence “Using the bottom-up emission inventory of CO₂ for the Los Angeles megacity and the CH₄:CO₂ ratio observed by the CLARS-FTS, we derived the CH₄ emission inventory using Eq. (3), where $E_{\text{CH}_4 | \text{top-down}}$ is the top-down CH₄ emissions inferred by the CLARS-FTS observations, $E_{\text{CO}_2 | \text{bottom-up}}$ is the bottom-up CO₂ emissions, $X_{\text{CH}_4} / X_{\text{CO}_2 | \text{slope}}$ is the $X_{\text{CH}_4(\text{XS})} / X_{\text{CO}_2(\text{XS})}$ ratio observed by the FTS and $M_{\text{CH}_4} / M_{\text{CO}_2}$ is the ratio of molecular weight of CO₂ and CH₄.” to “Using the bottom-up emission inventory of CO₂ for the Los Angeles megacity (166 ± 23 Tg CO₂ year⁻¹) and the average CH₄:CO₂ ratio observed by the CLARS-FTS (6.4 ± 0.5 ppb CH₄ (ppmCO₂)⁻¹), we derived the CH₄ emission inventory using Eq. (3), where $E_{\text{CH}_4 | \text{top-down}}$ is the top-down CH₄ emissions inferred by the CLARS-FTS observations, $E_{\text{CO}_2 | \text{bottom-up}}$ is the bottom-up CO₂ emissions, $X_{\text{CH}_4} / X_{\text{CO}_2 | \text{slope}}$ is the average $X_{\text{CH}_4(\text{XS})} / X_{\text{CO}_2(\text{XS})}$ ratio observed by the FTS and $M_{\text{CH}_4} / M_{\text{CO}_2}$ is the ratio of molecular weight of CO₂ and CH₄ (that is, $16 \text{ g CH}_4 / 44 \text{ g CO}_2$).”

13. pg 17052, lines 14-20. The statements concerning spatial variation in $X_{\text{CH}_4(\text{XS})} / X_{\text{CO}_2(\text{XS})}$ slopes suggests uncertainties are likely greater than estimated from Eq (3). It would appear more appropriate to state a range of CH₄ 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 $X_{\text{CH}_4(\text{XS})} / X_{\text{CO}_2(\text{XS})}$ 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 spatio-temporal 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 $X_{\text{CH}_4(\text{XS})} / X_{\text{CO}_2(\text{XS})}$ slopes observed for the 28 reflection points and defined the uncertainty of the average $X_{\text{CH}_4(\text{XS})} / X_{\text{CO}_2(\text{XS})}$ slope for the Los Angeles basin as the standard deviation among the $X_{\text{CH}_4(\text{XS})} / X_{\text{CO}_2(\text{XS})}$ 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 CH₄ 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 CH₄:CO slopes and CH₄ emissions. Is new data being reported from the work of Hsu et al (2009) and here in Table 4?

Response: There are two CH₄ emission results reported by Wunch et al. (2009). 0.40±0.10 Tg CH₄/ year is the top-down CH₄ emission estimated based on their CH₄:CO ratios while 0.60±0.10 Tg CH₄/ year is the emission estimated based on their CH₄:CO₂ ratios. We have clarified this in the caption of Table 4 by adding the following sentence “Wunch et al. (2009) reported two top-down CH₄ estimates: 0.40±0.10 Tg CH₄/ year derived from CH₄:CO₂ ratio and 0.60±0.10 Tg CH₄/ year derived from CH₄:CO ratio.” This can be found on page 25 of the revised manuscript.

The CH₄:CO₂ 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. Please see page 30 of the revised manuscript for the updated Figure 5.