

## Response to Anonymous Referee #1

We would like to thank the reviewer for taking time to review our paper and provide us with helpful comments. Please find our response to the reviewer's comments in blue in the following.

### General comments:

Wong et al. report measurements of CO<sub>2</sub> and CH<sub>4</sub> between 2011 and 2015 from a remote sensing instrument located on Mt. Wilson near Los Angeles, California, USA. Column CO<sub>2</sub> and CH<sub>4</sub> measurements above the instrument are subtracted from measurements from 28 points around the Los Angeles Basin in order to obtain an excess column enhancement below the instrument. These enhancements are fit linearly to provide excess CH<sub>4</sub> to CO<sub>2</sub> ratios, which are compared on a monthly basis to the other monthly measurements, and to previous studies. The ratios are also multiplied by CO<sub>2</sub> emission estimates from the basin to provide a monthly CH<sub>4</sub> emission. CH<sub>4</sub> emissions generally peaked in the late summer/early fall and wintertime in the Los Angeles Basin.

Overall, this paper is well-written and needs only minor revisions. However, the conclusions are seemingly at odds with another recent ACPD submission, Wunch et al. Whereas Wong et al. find Los Angeles Basin CH<sub>4</sub> emissions decreasing over the 2011 to 2015 time period, Wunch et al. report increasing CH<sub>4</sub> emissions from 2012 to 2015. I therefore suggest some discussion of the Wunch et al. results.

Response: Thank you for the reviewer's nice comments. Regarding the trend of annual CH<sub>4</sub> emissions over the 2011 to 2015 time period, our study concluded that there was no statistically significant trend in annual total CH<sub>4</sub> emissions over the 2011 to 2015 time period. In the abstract, the last sentence stated "The estimated annual methane emissions did not show a statistically significant trend over the 2011 to 2015 time period". Wunch et al. reported annual methane emissions from  $380 \pm 78$  Gg CH<sub>4</sub>/yr,  $352 \pm 71$  Gg CH<sub>4</sub>/yr and  $448 \pm 91$  Gg CH<sub>4</sub>/yr for the period 9/2012–8/2013, 9/2013–8/2014 and 9/2014–8/2015 respectively. It seems that there is not a significant trend in the emissions because the uncertainties overlap. Because Wunch et al. (2016) is still currently under review and has not been published, we did not cite their results in our paper. If their study is published, we would be happy to include a discussion of their results in our paper.

### Specific Comments:

1. Although there are many references to Wong et al. (2015), an example correlation plot of XCH<sub>4</sub>:XCO<sub>2</sub> would be useful in the current work.

Response: Thanks for the suggestion. We have added the following figure as an example correlation plot of XCH<sub>4</sub>:XCO<sub>2</sub> excess in the supplemental material.

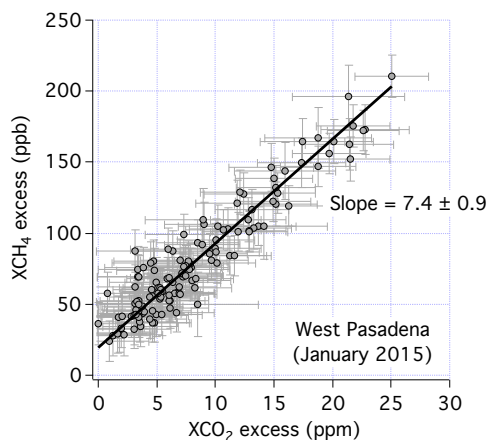


Figure S1. Scatter plot showing an example of correlation between XCH<sub>4</sub> excess and XCO<sub>2</sub> excess for CLARS-FTS west Pasadena target in January 2015. Regression slope of 7.4±0.9 was observed during this period.

2. p.14, line 7, the authors state Hsu et al. or Wennberg et al. showed wastewater treatment was responsible for the emissions stated. Both reported inventory values for wastewater treatment, but could not verify those inventories were correct. Since the definition of “showed” could be either “proved” or just simply “presented”, the authors should clarify this statement.

Response: We have clarified this by changing the word “showed” to “suggested”. The sentence (now at line 24 page 14) is updated to “This sector is suggested to be responsible for 33% of Los Angeles County and 9.4% of the South Coast Air Basin (Hsu et al., 2010; Wennberg et al., 2012)”.

3. Figure 5, I am curious what it would look like if average daily emissions were shown per month. As presented, the months with 31 days always seem to have small peaks compared to the surrounding months with less than 31 days.

Response: This is a good suggestion. We looked into that and found that the seasonal cycle does not have a significant change. Therefore, we decide to keep the units as total emission per month.

#### Technical Comments:

1. p.4, line 4, capitalize Transform Spectrometer

Response: The words “transform spectrometer” in “Fourier transform spectrometer” should be in lower case. Therefore, we did not make this change in our text.

2. p.4, line 6, remove comma between “to address”

Response: The change has been made in the text.

3. p.4, line 26, change to “molecules”

Response: The change has been made in the text. Please refer to line 2 on page 5 of the revised manuscript.

4. p.7, line 18, change ratio to MWCO<sub>2</sub>/MWCH<sub>4</sub> to match equation

Response: Changes have been made to the text. The sentence (now at line 22 of page 7) is updated to “The bottom-up estimate of R, the CH<sub>4</sub>/CO<sub>2</sub> emission ratio, was calculated from Eq. (4), where  $E_{\text{CH}_4}^{\text{inventory}}$  is the downscaled CARB annual total CH<sub>4</sub> emissions,  $E_{\text{CO}_2}^{\text{inventory}}$  is the downscaled CARB annual total CO<sub>2</sub> emissions and  $\frac{\text{MW}_{\text{CO}_2}}{\text{MW}_{\text{CH}_4}}$  is the ratio of the molecular weights of CH<sub>4</sub> and CO<sub>2</sub> (that is  $\frac{44 \text{ g CO}_2/\text{mole}}{16 \text{ g CH}_4/\text{mole}}$ ).”

5. p.8, is there a peer-reviewed citation for Hestia?

Response: No, the Los Angeles Hestia project does not have a peer-reviewed citation yet. The best citation for this project is <http://hestia.project.asu.edu>, which we have included in the paper already.

6. p.9, line 21, move comma to read “calculations, and (3) . . .”

Response: The comma has been moved. The sentence (now at line 29 of page 9) has been updated to “The differences result from 1) emission calculation methods, 2) the underlying dataset used in the emission calculations, and 3) spatial modeling.”

7. References, add Peischl et al.

Response: Changes have made in the text. In the reference section (line 9 on page 20), Peischl et al. 2013 has been added as “Peischl, J., Ryerson, T. B., Brioude, J., Aikin, K. C., Andrews, A. E., Atlas, E., Blake, D., Daube, B. C., de Gouw, J. A., Dlugokencky, E., Frost, G. J., Gentner, D. R., Gilman, J. B., Goldstein, A. H., Harley, R. A., Holloway, J. S., Kofler, J., Kuster, W. C., Lang, P. M., Novelli, P. C., Santoni, G. W., Trainer, M., Wofsy, S. C., and Parrish, D. D.: 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, 2013.”

8. Figure 6, line 5, suggest changing dash to colon for CH<sub>4</sub>:CO<sub>2</sub> ratio

Response: Thanks for the suggestion. We used the colon in past for our previous publication but the ACP journal typesetting suggested the use of dash instead. Therefore, we decide to keep the dash.