## 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 $CH_4$ or $CO_2$ abundance? Along those lines, how would spatial variability in $CH_4$ or $CO_2$ in the atmosphere above 1.6 km affect the excess ratio?

   Response: We define the excess $CH_4$ and $CO_2$ 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 $CH_4$ and $CO_2$ excess. We do not expect this to impact the $CH_4$:$CO_2$ excess ratio in the Los Angeles basin. Because a residual boundary layer consists of air mass from the previous daytime boundary layer, the relative $CH_4$:$CO_2$ ratio in the air mass would still carry information about the emission ratio of $CH_4$:$CO_2$ 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 $CH_4$:$CO_2$ excess ratio for a two-year period.

   Spatial variation in $CO_2$ and $CH_4$ 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 be 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 ± 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."

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 $CO_2$ or $CH_4$ source, which fill the boundary layer. We clarified this by editing the sentence on page 17039 line 28 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, the following text has been included "Spatial variation in the atmospheric column of $CO_2$ and $CH_4$ above Mount Wilson is minimal and does not affect the $XCH_4$:$XCO_2$ excess ratio. Spatial variation in $CO_2$ and $CH_4$ 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 $XCH_4$:$XCO_2$ 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 "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)."

7. Table 4; Why not add the 2010 CalNex $CH_4$:$CO_2$ from Wennberg et al. (2012)?

   Response: We have added the 2010 CalNex $CH_4$:$CO_2$ ratio (0.66 ± 0.03 ppb $CH_4$/ ppb $CO_2$) and the reported $CH_4$ emission (0.44 ± 0.1 Tg $CH_4$/ year) from Wennberg et al. (2012) in Table 4.

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 $CH_4$ (ppm $CO_2$)$^{-1}$ (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 $CH_4$ (ppm $CO_2$)−1 (Wennberg et al., 2012; Peischl et al., 2013; S. Newman, personal communication, 2014; Y.-K. Hsu, personal communication, 2014)."

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

    Response: Correction has been made in the text as requested.