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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Carbon Dioxide and Methane Measurements from the Los Angeles Megacity Carbon Project: 1. Calibration, Urban Enhancements, and Uncertainty Estimates" *by* K. R. Verhulst et al.

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Response to Anonymous Referee #3 Comments

1. General Comments:

The authors would like to thank anonymous referee #3 for a thorough review and many insightful comments that improved the manuscript. In response to one of the broader general comments, we agree it does not make sense to have both an Appendix and Supplement. We decided to transfer all Appendix materials into the Supplement, with changes made throughout the text to reflect updated figure and table numbers. Another general comment related to our background estimation method. As discussed in our responses to the previous two reviews, we have performed additional tests related to this topic. In our response to Reviewer #2, we include a new supplementary figure (Figure S4), showing histograms of the 1 hour S.D. for the CO2 and CH4 observations from the three background sites (SCI, LJO and VIC). We also made some minor changes to the background selection criteria for the three LA regional background sites (also discussed here), which we believe significantly improved the quality of the background estimates. All details regarding the background data selection methods are now included in the Supplementary materials, Section 3. Below we discuss these and other specific comments in more detail. Our responses are arranged by section of the manuscript. The reviewer also noted a number of very helpful grammatical and stylistic edits that we have addressed. These are summarized at the end of our response.

2. Specific Comments:

2.1 Abstract and Introduction

Pg 2, Ln 13: We have modified the text in the Introduction to include a discussion of using both top-down and bottom-up methods together to evaluate emissions (see P.2, 2nd paragraph). We also added several references (including: Lauvuax et al., 2016; Asefi-Najafabady et al., 2014; and Gurney et al., 2005).

Pg 2, Ln 30-Pg 3 Ln 3: The sentence noted was removed.

Pg 3, Ln 7: The Los Angeles project was described as a “pilot” because this is one of the first projects of its kind. For clarity, we changed the word “pilot” to “testbed”.

Pg 3, Ln 14: We decided to use an estimate of 16.3 million residents for the SCB region (roughly 17,100 km2) based on the following reference: The California Almanac of Emissions and Air Quality - 2013 EditionÂăRep., California Air Resources Board, Sacramento, CA (CARB, 2014). Ultimately, the population estimate should take into account the land area. For example, in 2015, the “Combined Statistical Area” for the greater Los Angeles area was 18.7 million. Combined statistical area for the greater Los Angeles area is defined as the sum of population in the five SCB counties, and including the non-urban desert areas of Riverside and San Bernardino Counties. A quick check suggests that there's roughly 1 million people in the desert areas of Riverside and San Bernardino (Victor Valley (Victorville + Apple Valley + Hesperia): 400k, Coachella Valley: 400k, Imperial Valley: 200k). Therefore, the estimate we are using is slightly smaller than the "Combined Statistical Area" because it does not include the non-urban desert portions of the Los Angeles, Riverside and San Bernardino Counties in the SCB.

Pg 3, Ln 22: We agree. The redundant sentence was removed, with some other minor stylistic edits to the text on P. 3 lines 29-31 for clarity.

Pg 4, Ln 1-5: This comment relates to the ethanol content in gasoline used in California, which the ARB reports has been approximately 10% by volume since the beginning of 2010. Ethanol derived from C4 grasses (i.e. corn) increases the ratio of 13C/12C from combustion of gasoline, which adds complexity to fossil CO2 attribution. Newman et al., (2016) discuss this topic in great detail. We changed the text in the Introduction, P. 5 (∼lines 12-14) as follows: "In California, gasoline is approximately 10% ethanol by volume. Ethanol that is derived from biofuel (i.e. from C4 grasses, such as corn) will increase the ratio of atmospheric 13C/12C when gasoline is combusted, adding complexity to the attribution of fossil CO2 emissions (Djuricin et al., 2010; Newman et al., 2016)."

Pg 5, Ln 1: We updated the text to reflect these changes. On P. 5, we define the first instance of the term "enhancement."

Pg 5, Ln 2-3: The word "robust" was changed to "large" and "roughly" was removed in both instances. We kept "10s to 1000's of ppb CH4" because this still accurately reflects the range of CH4 mole fraction reported from multiple studies in the LA Basin.

2.2 Methods Section

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Pg 7, Ln 23: In our system, the temperature of the Earth Networks sample module is controlled at 38 degrees C at all sites. To address the second part of this comment, the Earth Networks sample module has a heater which is strong enough to raise the temperature in the box by up to about 30C. Therefore, the optimal range of control for the heater is when the ambient (room) temperature is between 8C to 38C. The GCWerks software controlling the sample module was modified to implement a PID algorithm to keep the temperature control to within about one tenth of a degree under normal operating conditions. In GCWerks, we also monitor the variables “sample temperature,” which is the readout of the sample module temperature (after control), and “ambient temperature, which is the temperature immediately outside the sample module. These variables can be used to look at the stability of the sample module and room temperature over time. As the reviewer inferred, we do not actively cool the sample module and only apply heating. Therefore, we rely on the ambient air temperature just outside of the sample module to be within the 8C to 38C range so that the only primary cooling mechanism required is the loss of heat from the sample module to the room. While this could introduce potential problems during an extreme summer heatwave, the majority of the instruments are located in temperature controlled shelters so the ambient room temperature is nearly always between 8C and 38C. To address this concern, we included the temperature at which the heated box is maintained (38C) in the text on P. 7 (line 30).

Pg 9, Ln 19-20: The high mole fraction tanks were purchased from NOAA/ESRL. The tank filling procedure for all tanks (near ambient and high mole fraction) is now described in the first paragraph in Section 2.4. For reference, the NOAA/ESRL tank filling procedures are described in much more detail here: https://www.esrl.noaa.gov/gmd/outreach/behind_the_scenes/standards.html. The text on P. 9 (lines 12-18) was changed as follows: “In addition to the ambient-level calibration and target tanks, the VIC and LJO sites had high mole-fraction standard tanks installed at the time of this study. These high mole fraction tanks were prepared by NOAA/ESRL and calibration assignments were provided prior to deployment (roughly

500 ppm CO2 and 2600 ppb CH4). The NOAA/ESRL high mole fraction tanks are prepared by adding a 10% CO2-in-air mixture to natural air during the pressurization of the cylinder at Niwot Ridge, Colorado (and a similar procedure is used for CH4). The cylinder is then moved to the NOAA calibration laboratory in Boulder, CO where it is calibrated relative to NOAA/WMO secondary standards."

Pg 9, Ln 20-23: Yes, we reject the first 10-minutes to account for the stabilization period after the inlet is switched, i.e. to account for the turnover of air in the CRDS coming to equilibrium (see also Welp et al. 2013). This was mentioned in the supplement, however, we also modified the main text for clarity (now on P. 10, lines 2-4): "The first 10 minutes of each tank run are rejected and only the data from the last 10 minutes of any are used in the calibration of CO2 and CH4 mole fractions to account for the stabilization of air in the CRDS after the inlet is switched (Welp et al., 2013)."

Pg 9, Ln 14: Yes, we are evaluating both accuracy and stability, where stability is the accuracy over time. We added a sentence on P.9 (line 21), as follows: "This calibration method assumes a linear response in the analyzer."

2.3 Results Section

Pg 10, Ln 23-24: We decided to remove the first sentence of Section 3.1 and made some minor revisions to the text of the paragraph that follows to begin this section. The next sentence was revised as follows: "Figure 2 shows the 1 hour average observations collected from nine sites in the Los Angeles surface network between January 1, 2013 and December 31, 2015 and Tables 2 and 3 show the annual average CO2 and CH4 variability based on hourly observations collected during 2015."

Pg 11, In 1: We included the word "hourly" to clarify that the S.D. is defined using the hourly data. We also modified the Table 2 and 3 captions so it is clear that we using hourly data to compute the annual statistics.

Pg 11, Ln 13-14: Yes, the VIC and SCI sites exhibit the lowest variability with regards

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to CO2 and CH4 mole fractions. The text was modified slightly to clarify this point. We changed the text in the second paragraph of section 4.1 as follows: “Victorville and San Clemente Island (VIC and SCI) show less variability in CO2 and CH4 mole fractions compared to the other sites within the SCB (Figure 2).”

Pg 12, Ln 14-15: Yes, the assumption was implied. We modified the text so this is now explicitly stated on P.12 (now on lines 18-20) as follows: “Given a constant flux, and assuming that transport in and out of the boundary layer remains approximately constant, the trace gas mole fraction observed within the PBL will increase or decrease as the PBL height falls or rises, respectively.”

Pg 12, Ln 17-18: We agree, the PBL height, rather than stability, is the more important factor controlling the magnitude of the enhancements. A similar comment was made by reviewer #1 in the same section of the text, so we have modified the text on P. 12 to address both comments, see lines 14-15: “Shallower PBL heights at night will lead to higher trace gas enhancements and higher sensitivity to local surface emissions (e.g., Djuricin et al., 2010; Turnbull et al., 2015).”. . .and lines 23-24: “The reduced variability in the CO2 and CH4 observations during midday hours is in part due to the larger height of the PBL during the mid/late afternoon.”

Pg 13, Ln 10: A consensus on the notation for CO2 and CH4 enhancements is indeed a good question for the broader community and perhaps something that should be discussed as part of upcoming workshops that include urban measurements. The terms enhancement and excess are interchangeable and we agree with the reviewer that the capital “delta” notation could be confusing to some readers. We decided to adopt the notation “CO2xs” in alignment with the work of Newman et al. (2013; 2016). All instances of “delta” notation were removed and were changed to CO2xs or CH4xs (including figures, tables, and captions).

Pg 13, Ln 18: This is a minor technical comment. We decided to leave the units in for clarity. Sometimes readers may not start with the beginning of a paper, so we feel it is important to note that we are using the mole fraction and indicate the units used throughout the paper in this section.

Pg 13, Ln 19: This is a stylistic point. We have decided to leave the title of Section 4 as is as it describes our process for evaluating background mole fractions for the Los Angeles region (as “estimation” is the process of approximately calculating or evaluating something.

Pg 13, Ln 22-25 and Pg 14, Ln 15-16: We also decided to leave in the site details in these locations for clarity. We also added the site codes for the LJO and PVP sites.

Pg 15, Ln 23: Correct, MWO is a mountaintop site and “m asl” is the correct notation!

Pg 16, Ln 13-14; Pg 16, Ln 25-26; Pg 16, Ln 27-28; and Pg 17, Ln 7-10: Here we respond to four comments all related to Figure 4, which show the MBL curves and a comparison of the background estimates. In Figure 4 we show 3 MBL curves, one near the latitude of SCB and the other two from latitudes north of the Basin. Generally, the climatological flow into Southern California during onshore flow conditions is from the north rather than the south, before reaching the California Bight and flowing inland. This is illustrated in the HYSPLIT back trajectories shown in Figure 5, and is the primary reason we chose to show MBL estimates for two latitudes north of the SCB region. Aside from this, the exact latitudes for the MBL curves are linked to model resolution. Another related comment in this set refers to the lower panels of Figure 4, where we show the difference of each smooth curve estimate from a “control” case. In the manuscript, this difference was computed from the MBL curve at 40.5 N. Overall, the differences in the CO2 and CH4 mole fractions between the three MBL curves at 40.5, 36.9 and 33.4 degrees N are relatively small (see Figure 4, upper panels). Therefore, the choice of subtracting each curve from the 40.5N latitude was somewhat arbitrary. Per the reviewer’s suggestion, we decided to subtract each curve from the MBL estimate at 33.4 N to update the analysis. In response to the last comment in this set, the estimates for the background uncertainty were computed as the “percentage of the enhancement" for mid-afternoon hours only.

Summary of MS changes related to these comments: We updated Figure 4 and modified the text in Section 4.2 (P. 16-17) to indicate that we subtracted each background estimate from the MBL curve at 33.4 N: "The average absolute difference between the Pacific MBL estimate at 33.4° N and each background estimate from SCI, LJO, VIC, and MWO for the period shown in Figure 4 is: 0.8, 0.7, 1.7 and 1.5 ppm CO2, and 8.0, 8.9, 10.1, and 13.7 ppb CH4, respectively. The average absolute differences between the background estimates from SCI and LJO and the Pacific MBL estimate from 33.4° N are <1 ppm CO2 and <10 ppb CH4, suggesting that both sites are useful for deriving marine background estimates for CO2 and CH4 when the appropriate filtering criteria are used." We also updated the text in Section 6.2 (P. 28, ∼lines 16-18): "During 2015, the annual average uncertainty in the SCI smooth curve estimate is 1.4 ppm CO2 and 11.9 ppb CH4 (Table 6). This amounts to roughly 10% and 15% of the median mid-afternoon enhancement in Downtown LA (USC) for CO2 and CH4, respectively." And in the Abstract (P. 2, ∼lines 3-4): The background uncertainty for the marine background estimate is ∼10% and ∼15% of the mid-afternoon enhancement near Downtown LA for CO2 and CH4, respectively.

Pg 17, Ln 2-5: LJO is a coastal site with essentially no local upwind sources over southwesterly sector. Generally, the CO2 and CH4 observations from LJO show significantly more variability than the San Clemente Island and Victorville sites. As discussed in the manuscript, the variability at LJO is more like an urban/suburban site than a background site. This is primarily because the site has very strong sources in other wind sectors (and the reason we see measurements as high as 5ppm, which is indeed very high for a background site). The proximity of the LJO site to local sources (including a large landfill immediately to the east and along-shore transport from the north) as well as the meteorology (which sometimes brings very clean air to the site) explains why this site is both heavily impacted by local emissions but is sometimes useful as a background site. Regarding the variability in both species, CO2 and CH4 can be coemitted from urban emissions sources, such as landfills, gas fired power plants, etc. Therefore, both CO2 and CH4 levels (as well as CO and other trace gases) may be impacted when the site is influenced by an urban air mass. By requiring small variability in both CO2 and CH4 levels as part of our filtering criteria, we are assuming that either gas could indicate influence from an urban air mass. While we did not find that the selection criterion based on both CO2 and CH4 variability was too strict, we did make small modifications to the data selection criteria as described in our response to comments from Reviewer #2 as well as the next comment. For further changes related to the background topic, please see our response to the comment below as well as our Response to Comments from Referee #2.

Pg 17, Ln 10: We agree, Mauna Loa is a very good background site and our criteria for the LA background sites need not be identical to this site. Based on reviewer comments and our internal review, we performed several additional tests with the background selection criteria. In our response to Reviewer #2, we included a new figure in the Supplement (Figure S4) with histograms of the 1 hour S.D. for the CO2 and CH4 observations from SCI, LJO and VIC, similar to the analysis of Thoning et al.. During 2015, 70%, 42%, and 30% of the data had a 1 hour S.D. <0.3 ppm CO2, 67%, 57%, and 42% of the data had a 1 hour S.D <3 ppb CH4 filter criteria, and 60%, 35%, and 29% of the data met both criteria for the SCI, VIC, and LJO sites, respectively. Based on this analysis, we reduced our CH4 one hour S.D. filter limit from 5 ppb to 3 ppb CH4 and retained the original CO2 filter criteria (0.3 ppm CO2). Next, we performed tests by varying the number of consecutive hours with stable conditions from 3 to 6 hours and the cutoff for the hour-to-hour variability between 0.25 to 0.5 ppm CO2. We found that the LJO and VIC observations were most sensitive to the filter parameters, especially criteria 2 and 3, the hour-to-hour stability and number of consecutive hours. For VIC, our results are in agreement with Reviewer #3's inference that the limits were too strict. We found that requiring 6 or more consecutive hours of stable conditions at VIC resulted in large data gaps over the entire season during summer months, making the background estimate highly uncertain during this period. We also found that gaps in the VIC background observations were reduced to <1 month when the following criteria were used: 1) hour-to-hour stability equal to 0.5 ppm CO2 and 2) number of consecutive hours with stable conditions equal to four hours. For LJO, the original filter criteria did not produce gaps >1 month during 2015-2016. Furthermore, using less strict criteria for LJO resulted in a few anomalously high CO2 and/or CH4 observations being included in the result, which is unfavorable (and likely due to a persistent polluted air mass passing over the site rather than clean background air). For these reasons, we decided to keep the same filter criteria for both LJO and SCI. With these changes, there are no longer significant gaps in the records used to generate the smooth curve fits. Overall, we believe the results presented are now very reasonable, as exhibited by the improved agreement between the background estimates. Finally, we note that while the filtering criteria are now less strict, there are still differences between the marine (i.e. LJO and SCI) and continental (i.e. MWO and VIC) background estimates that cannot be explained by the data filtering methods.

Summary of MS changes for this comment: Supplementary materials: See Figure S4 showing histograms of the 1 hour S.D. for the CO2 and CH4 observations from SCI, LJO and VIC. Also, see new text detailing the filtering criteria in Section 3 (P. 4-5). In Section 4.1 of the main text, we now refer to details about the filtering criteria in the Supplement. In Section 4.2, P. 17 (∼lines 3-6), we modified the text as follows: “The cause of the larger differences between the continental (i.e., VIC and MWO) and marine (i.e.. SCI, LJO, and Pacific MBL) background estimates is not clear. Future modelling studies could investigate whether a time-dependent background selection method – e.g., based meteorological information and the origin of incoming air mass – can be used to determine the appropriate background site under some of the more common meteorological regimes in the SCB.” We also removed the following sentence that was no longer relevant in Section 4.3, P. 18: “For VIC, there is virtually no CO2 or CH4 data meeting the selection criteria during the summer and early fall months (Figure 3).”

Pg 17, Ln 18: Yes, the back trajectories are for the Caltech site in Pasadena, CA using NAM12 winds. We modified the text in Section 4.3, P. 17 (line 17-18) as follows: “We computed twenty-four hour back trajectories for winds arriving at the CIT site in Pasadena at 14:00 LST using NOAA’s HYSPLIT model (Figure 5; Stein et al., 2015; Rolph, 2016).” We also added more details to the Figure 5 caption, including the site coordinates, the model (HYSPLIT) and that we used hourly winds from NAM12.

Pg 17, Ln 32: As noted in our response to an earlier comment, all the background curves in Figure 4 are now subtracted from the MBL estimate at 33.4 degrees N. Figure 5 is mainly intended to show the general seasonal trends in the location of the incoming air masses to the SCB region. We mainly showed the 32.5-36 N latitude range because expanding the scale did not add much additional information and the zoomed-out view made visualizing the back trajectories more difficult. However, we note that this analysis has also been performed for other sites in the SCB and shows very similar results.

Pg 19, Ln 19: We modified the text to clarify that observations shown in Figure 6 are averaged for 2015, as opposed to the whole record.

Pg 19, Ln 22-28: We removed some of the redundant text and merged the remaining text from the first and second paragraphs of Section 5, which significantly improved the overall flow of this section.

Pg 20, Ln 2-3: The main issue noted relates to seasonal variations in mixing layer height. We prefer to cite the Ware et al., (2016) rather than Strong et al., (2011) for relevant information on mixing heights in Los Angeles and because winter conditions may impact mixing heights differently in LA and Salt Lake City. Recently, Ware et al., (2016) used backscatter data from a Mini Micropulse Lidar (MiniMPL) instrument located at Caltech in Pasadena, CA and provided a detailed assessment of mixing height observations from 2012-2014 near one of our rooftop sites. Their results suggest relationship between trace gas enhancements and mixing height in LA may be more complicated than the reviewer's comment suggests. We added text to the manuscript on P. 20 (lines 11-21), as follows: "In general, increased summertime insolation is expected to produce a deeper afternoon mixed layer depth in summer relative to winter, which in turn would result in larger trace gas enhancements within the PBL during winter relative to summer. As discussed earlier, Ware et al. (2016) used backscatter data from a Min-iMPL instrument located in Pasadena, CA to estimate mixing heights over two years from 2012-2014. They found the mean afternoon maximum mixing depth was 770 m agl in summer (June and August) and 670 m agl in winter (December–February). However, seasonal differences in mixing depth should also be considered in the context of the daily and weekly variability. Ware et al. (2016) show that the maximum depth of the afternoon mixing layer may differ by a factor of 2 from day-to-day. Furthermore, they also show that the within-season S.D. for the afternoon maximum mixing height is about 220 m, or approximately 30% of the mean afternoon maximum mixing depth in either summer or winter (which is larger than the observed average seasonal differences in mixing height). Overall, the large variability in mixing layer depth over different timescales suggests that the meteorological impacts on trace gas concentrations in the PBL can also be quite variable."

Pg 20, Ln 11: The word "outlier" was used incorrectly here and in the figure caption. The red pluses in Figure S11 indicate enhancements greater (or less than) the maximum whisker length, showing the full range of variability.

2.4 Summary and Conclusions Section

Pg 30, Ln 17: We are interested in the magnitude of the enhancement, which is the signal above background, not the signal relative to the enhancement. A minor modification was made to the text to make sure this point is clear

Pg 31, Ln 20: The run-on sentence was rewritten and the instances of the phrase "we plan to" were also revised as suggested.

2.5 Appendix/Supplement

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Appendix A1: Pg 56, ln 13-17: Please note, all appendix materials are now included in the supplement, so the text noted here is in now located in Section 1.2 of the Supplementary materials. In general, manual flags are applied on a case-by-case basis. Mainly, the manually flagged data are identified during technician site visits, especially those that required modifications to the plumbing on the instrument of callbox (e.g. when calibration standards are replaced, when an analyzer is removed for repair, etc.). The text was modified to describe some specific instances when manual flags have been applied.

3. Typographical edits

All suggested typographical edits listed below were addressed in the revised manuscript:

Abstract Ln 30: The time span should be explicitly stated, not “roughly”. This can be fixed by removing the word “roughly”.

Pg 3, Ln 7: You don’t need to cite the URL a second time since it was listed on the prior page.

Pg 6, Ln 19-20: missing the word ‘to’ here: “. . .were often inaccessible due TO permitting or other restrictions.”

Pg 9, Ln 17: Forgot a period at the end of the sentence.

Pg 10, ln 28: “levels” should be “mole fractions” for better clarity.

Pg 11, ln 1: You don’t need the “roughly” since you define the time range and are using hourly data, unless the hourly window changes from day to day.

Pg 12, Ln 11: “. . .controlling the variability of CO2 (and CH4) observations. . .” should be “. . .controlling the variability of TRACE GAS observations. . .”

Pg 12, Ln 31: Again, “CO2 (and CH4)” should be “trace gas.” Wind speed affects other trace gases as well (CO, NOx, etc).

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Pg 15, Ln 9: The word “very” in unnecessary.

Pg 15, Ln 11-12: I wouldn’t describe the selection criteria used by Thoning et al 1989 as “preliminary,” its just what they used.

Pg 16, Ln 1: MBL should be defined as Marine Boundary Layer.

Pg 16, Ln 13: “roughly” is not needed since the exact latitudes are listed.

Pg 17, Ln 13: The “LA Basin” should probably be SCB for consistency.

Pg 18, Ln 20: There is an extra parenthesis next to Feng 2016.

Pg 22, Ln 4: There is an extra “and” on this line.

Pg 30, Ln 7-10: Way too many instances of the word “roughly” in this paragraph. Roughly is even duplicated on line 10! The authors could simply remove every instance of “roughly” and it would read better, or they could simply report the specific values and that would be fine also.

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