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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.

Anonymous Referee #3

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General Comments

This manuscript describes the instrumentation and calibration methods used for the Los Angeles Megacity surface observation network. The authors present a method for calculating background mole fraction conditions using sites outside the domain of the South Coast Air Basin and utilize this background to calculate urban enhancements above background. Finally, they describe the components of the analytical uncertainty that affect the observations. In general the manuscript is well written and the experimental methods are described in detail. This is an important methodological description of a major project and is therefore an important contribution and within the scope of Atmospheric Chemistry and Physics.

While I have a few suggestions regarding the background selection criteria and numerous technical suggestions, the bulk of the manuscript is technically sound and I would recommend publication after these minor concerns are addressed.

One complaint is that there is an Appendix A1-3 as well as Supplemental Materials. It would simplify the manuscript if the authors changed the supplemental information to an Appendix B, or if they added Appendix A1-3 to the Supplemental Materials. It seems overly complicated to have both an Appendix and supplemental information.

Specific Comments

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

Pg 2, Ln 13: “Carbon fluxes can be estimated using “top-down” or “bottom-up” methods.” – Why use “or” in this statement? It is possible to use both top-down and bottom-up methods to evaluate emissions, and it is also possible to use both methods in an iterative process that leads to an optimized solution. I think a discussion of how these methods can be used together is missing from this paragraph.

Pg 2, Ln 30-Pg 3 Ln 3: The statements starting with “More information flow. . .” seems like it doesn’t belong in the introduction since its making a recommendation. Looking at the conclusions, this information is there prominently and that is what it should be so readers pay attention to it, but I don’t think you need to also have the recommendation in the introduction. It would be fine to describe how methods should be documented and how there is a long latency in most observational networks currently (etc) since that is the current state of affairs, but leave the recommendations for the discussion/conclusions (in my opinion).

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

Pg 3, Ln 7: Is the LA project considered a “pilot” project? What would a full blown project be if the LA project is only a “pilot”?

Pg 3, Ln 14: Is there a more accurate population estimate than “> 15 million”? Could it be 20 million? What does the US Census Bureau say?

Pg 3, Ln 22: The sentence starting with “Urban and suburban areas. . .” seems redundant to the next paragraph where this is discussed in greater detail. I seems like you could just remove this sentence and have this paragraph describe the met conditions and the topography, while the next paragraph describes possible emission sources & complexities. Pg 4, Ln 1-5: You should also list the complication of using ethanol in gasoline. This will have a different d13C signature than fossil gas because its derived from C4 grass (corn) and is an additional important complication for understanding CO2 emissions.

Pg 5, Ln 1: “Enhancements” should be defined when you first use it. Also, “robust” seems to be an odd descriptor. Perhaps “large enhancements” would be better?

Pg 5, Ln 2-3: The word “roughly” is redundant to listing a range of mole fractions. Also, for CH4 surface mole fractions it would be better to list a range like was done for CO2 instead of saying “10’s to 100’s of ppb” in order to keep the text internally consistent.

Pg 5, Ln 31: The word “extensible” seems odd. Would it be better to say “. . .are intended to PROVIDE A BLUEPRINT FOR other surface observation. . .”? Also, don’t you provide recommendations about how to improve your calibration setup? From the conclusions: “In the near future, the LA measurement network will begin using analyzer specific estimates of the correction factor based on periodic measurements with high mole fraction tanks, which will allow correction of the random and systematic components of the uncertainty associated with the single-point calibration strategy.” It might be nice to mention that you will be providing suggestions for future deployments as well here.

Methods: Pg 6, Ln 19-20: missing the word 'to' here: "...were often inaccessible due TO permitting or other restrictions."

Pg 7, Ln 23: What temperature is the heated box kept at? Is it kept at a constant temperature (ie – is there a fan that cools it down when its too hot) or is there just a heater for when it is cold?

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

Pg 9, Ln 19-20: I think this is the first reference to high mole fraction tanks, but the filling procedure for these tanks has not been described. A brief description of how high mole fraction tanks are filled and calibrated is warranted, probably as part of the preceding paragraphs that talk about the filling and calibration of ambient mole fraction tanks.

Pg 9, Ln 20-23: I assume that the first 10 minutes of calibration data are rejected because that is longer than the turnover time of air in the CRDS and the instrument is coming to an equilibrium. If so, it would be nice to state this.

Pg 9, Ln 14: This calibration method assumes a linear response in the analyzer, and I think this is an important assumption to state clearly in the manuscript somewhere in this section. Also, I think this sentence is slightly misworded: "The sensitivity (S) of the high mole fraction tank is also tracked over time, providing a check on the analyzer stability at higher mole fractions." I don't think you are evaluating the "stability" but instead you are evaluating the "accuracy."

Pg 10, Ln 23-24: The first line of the Results is incomplete: "Atmospheric CO2 and CH4 mole fractions can vary on timescales ranging from less than 1 hour, to annual, and inter-annual cycles." Of course there can also be decadal, centennial, and millennial variations as well. Perhaps this could be changes to say that "The atm CO2 & CH4 in our data sets contain variations from less than one hour to annual and iner-annual", or

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something like that.

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 11, ln 1: Also, is the S.D. defined using the hourly data? Its clear that these statistics are for “afternoon hours,” but it is not clear that it is using hourly data (vs minute average data, or daily afternoon averages). This could easily be clarified in the text.

Pg 11, Ln 13-14: “We find that SCI and VIC are the cleanest sites in terms of their annual CO2 and CH4 variability.” – It is unclear what “cleanest” means in this context. Does it mean they have the lowest variability?

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

Pg 12, Ln 14-15: This brief description of a box model framework for how the PBL affects mixing ratios is missing the assumption that transport in and out of the boundary layer remains approximately constant.

Pg 12, Ln 17-18: I disagree with this statement: “The reduced variability in the CO2 and CH4 observations during midday hours is in part due to the stability of the PBL depth during the mid/late afternoon.” The stability is due to the larger size of the PBL during the middle of the day (compared to the night), not necessarily the “stability of the PBL depth.” Because the PBL is larger, a particular flux will be more dilute in a larger ‘box’ and therefore you’ll see lower variability. If my understanding is incorrect it would be good to have a citation here to back this claim up. After reading a few more lines I see this is mentioned on lines 22-24. However, I think the size of the PBL is a bigger factor than the “stability of the PBL depth.”

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 13, Ln 10: The use of capital delta CO2 or capital delta CH4 is slightly controversial. The problem is that the capital delta symbol is used to represent radiocarbon measurements, so its use to represent local enhancement could be confusing. The research community has not agreed on what the appropriate symbol to use to represent local enhancement of GHGs, so the authors should feel free to use what they feel is appropriate. However, they should be aware that this is something that should be agreed upon within the community and the authors should only use this symbol if they intentionally think this is how it should be displayed, because others will follow their example. Other options that I am aware of are writing out “CO2 enhancement” or “excess CO2” or “CO2-ex”.

Pg 13, Ln 18: I don’t know that you need to continually state that the units are ppm or ppb. Its clearly stated in multiple places before this.

Pg 13, Ln 19: The authors are calculating background mole fractions, not estimating them.

Pg 13, Ln 22-25: Listing the lat/lon is unnecessary since these sites are described in Table 1. Only list the lat/lon if the site is not already in Table 1. Also, the site codes should be used here instead of describing the sites.

Pg 14, Ln 15-16: Again, lat/lon in unnecessary and Table 1 should instead be cited.

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 15, Ln 23: “1670 m agl” should be “1670 m asl”, unless this site is on a VERY large tower. :-) The authors could also just refer to this as being on a mountaintop and refer to table 1 for the elevation.

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 16, Ln 13-14: The selection of these MBL latitudes deserves more explanation. Obviously 33.4N is the latitude of the SCB, so that makes sense. But why look at two other latitudes north of that, but not a latitude south of that? Also, 40.5N is ∼770km north of the SCB, so is this really a good comparison to make?

Pg 16, Ln 25-26: This is surprising to me since I don't know of a mechanism that would draw CO2 down over land during the winter. My guess is that this difference may have to do with how the 2-D MBL was computed (there is greater smoothing in the 2D MBL curves than in their background curves), or perhaps the latitude chosen. Looking at Fig 4 (top left) closely, the springtime MBL at 33.4 is closer to the authors background estimates than the one from 40.5N. Given this, I don't understand why the authors chose to subtract their background estimates from the 40.5N MBL in the bottom panels of Fig 4 instead of using the MBL from 33.4N, which is the latitude of the SCB. Either the authors should explain the rational for this selection or possibly change the latitude they are using in the bottom panels of Fig 4.

Pg 16, Ln 27-28: It looks to me like they are pretty similar in the summer of 2014, but very different from each other in the summer of 2015. Also, I have the same question/comment about using the MBL from 40.5N instead of 33.4N for CH4 as well.

Pg 17, Ln 2-5: Yes, the summertime CH4 from LJO is very different from the other sites! This is really clear in Fig 3, so there should be a reference to that figure in this section. In fig 3 it is clear that LJO has measurements as high as 5ppm which is very high for a background site. There are some very abrupt changes in the LJO data set that may indicate the start or end of a physical process (landfill being covered or not). By having the selection criteria reject observations if there is too much variability in both species they may be making their criteria overly selective at this site.

Pg 17, Ln 7-10: These would be slightly different if the 33.4N latitude was used. Also, it may be described later, but for the % enhancement figures it should be stated that

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this is during the afternoon (if that is the case).

Pg 17, Ln 10: After reading this section I went back and looked at Fig 3. It seems to me that the background selection criteria is too strict for VIC and LJO since there are large parts of the year when there are no selected data. This is problematic, for example, at VIC in the summer of 2015 when there are no selected data and the smooth CH4 curve dips below all of the data. The CCGCRV software relies on high and low pass filtering of the data to compute the smoothed curves, but if there is no data it doesn't work! Based on this, I would recommend that the authors slightly relax their selection criteria so that they have at least some data throughout the year. Otherwise they are interpreting a smoothed curve that isn't based on anything. Another approach could be to mask out their background during times when they don't have data to constrain the smoothed curve, but that would defeat their objective of having a background curve all the time. Looking back at Pg 15, Ln 11-21 made me look again at Thoning et al 1998, and they plotted the distribution of their data at Mauna Loa in their Fig 1 showing that most of their data had an hourly variability <0.3 ppm. I wonder what it would look like if the authors examined a similar plot for their data? My guess is that there is greater variability in the data in this work than there was at Mauna Loa (Mauna Loa is a very good background site!) and perhaps a less stringent selection criteria is needed so that they can have more data present throughout the year. After reading further, this issue is discussed on pg 18 ln 1-10, but the authors conclude that VIC is just not suitable as a background station during the summertime due to onshore flow. In that case, the smoothed VIC background curve shouldn't be discussed. However, I still feel that there could be a compelling case to make the selection criteria less stringent than that used for Mauna Loa.

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

Pg 17, Ln 18: Did you select a specific site in Pasadena? It might be appropriate to list the lat/lon on this location (even though the specifics don't really matter that much). If the back trajectories originate from a site, that would be ideal to note. Also, what meteorology did they use to drive the HYSPLIT model? The authors state they were using trajectories that ended at 14:00 PST, but were they using monthly averaged wind fields, or were they selecting meteorology from representative days? This should be explicitly described because the decisions about met fields would affect the back trajectories greatly, and hence the interpretation of the seasonal patterns of dominant transport.

Pg 17, Ln 32: The plot showing the back trajectories only spans from 32.5N to 36N, so this seems to also question why the authors looked at MBLs extending all the way up to 40N.

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

Pg 19, Ln 19: The sentence starting "Figure 6 shows. . ." should explicitly state that the data is averaged for 2015 (as opposed to the whole record). This is stated in the Figure caption, but its ambiguous in the text.

Pg 19, Ln 22-28: These two paragraphs are redundant to Table 4 & 5 and could be removed.

Pg 20, Ln 2-3: The sentence starting "Prior studies. . ." should have citations with it. Also another factor in the greater wintertime enhancements is the lower PBL heights, as shown in Strong et al. (2011), Figure 4a.

Strong, C., C. Stwertka, D. R. Bowling, B. B. Stephens, and J. R. Ehleringer. "Urban Carbon Dioxide Cycles within the Salt Lake Valley: A Multiple-Box Model Validated by Observations." Journal of Geophysical Research: Atmospheres 116, no. D15 (2011): n/a–n/a. doi:10.1029/2011JD015693.

Pg 20, Ln 11: How were the "outliers" defined? Often outliers are rejected, but it seems that none of these were? It would be good to state this explicitly if the authors use the "outlier" terminology.

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.

Pg 30, Ln 17: "enhancement" is already defined and used in prior paragraphs, so this can just be stated like this "...and how large the signals is relative to the enhancement."

Pg 31, Ln 20 – The sentence starting with "As part of future work..." is a run-on sentence that is a bit confusing, so it should be re-written. Also, I have a small quibble with using the phrase "we plan to..." here and in many other places in the manuscript (eg Pg 19, Ln 3; pg 26 Ln 26, etc). I think that writing "we plan do this or that" lays claim to doing work in the future and discourages others from examining work that needs to be done. Instead, it would be better to say "this or that should be done in the future." This is because investigators change, authors change, plans change, collaborators change, etc. What if another research group comes up with a new method and you collaborate with them to examine fluxes? This could be re-written as: "As part of future work, forward and inverse modelling studies as well as tracer-tracer analysis SHOULD BE USED TO...", or something like that depending on how the run-on sentence is re-worded.

Appendix A1: Pg 56, ln 13-17: The text reads: "Data flags are applied by Earth Networks based on recommendations from the LA Megacity Data Working Group, a team of scientists from NASA's Jet Propulsion Laboratory, 15 Scripps Institution of Oceanography, National Institute of Standards and Technology, and Earth Networks. The manual flags are applied on the EN server to indicate those data that are not recommended for further scientific evaluation or interpretation within the scope of the project." It would be useful for other research groups working on urban GHG measurements to describe what these manual data flags are, or provide a citation for the conclusions of this working group. This is a very important addition.

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