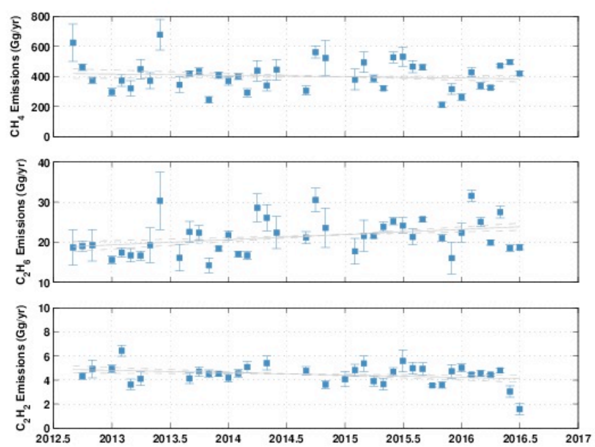
Response to Anonymous Referee #2.

We thank the referee for their valuable comments, which substantially improved the paper.

Referee comments are in ***red italics***, our responses are in black text.

The authors should consider and discuss the statistical significance of the reported trends in observed C1 and C2. The confidence intervals around the annual averages in Table 1, for example, suggest the annual averages across the 3-years shown are not statistically different. On the other hand, assuming the error bars shown in Figure 3 are correct, the 2015 values for C2 emissions seem to be statistically higher than those during 2006-2010. The authors should consider whether the monthly C1 and C2 emission time series in Figure 6 provide an alternative basis to determine the existence of a significant trend (e.g., are the slopes statistically different than zero?).

We have computed slopes for the monthly emissions. There is no statistically significant trend in the methane emissions during the 2012-2016 period (-9+/-14Gg/yr), and a very slight decrease in acetylene (-0.20+/-0.15Gg/yr). There is a statistically significant increase in the ethane emissions during this period of (1.3+/-0.6Gg/yr). We also looked back at data from two other temporary TCCON stations in the SoCAB (2007-2008 and 2011-2013) for which we can compute methane emissions (but not ethane



or acetylene). Between 2007-2015, there is a (very) slight decrease in methane emissions (-5+/-4 Gg/yr), which is in good agreement with the Wong et al. (2016) estimate of -5+/-4 Gg/yr.

Wong, K. W., T. J. Pongetti, T. Oda, P. Rao, K. R. Gurney, S. Newman, R. M. Duren, C. E. Miller, Y. L. Yung, and S. P. Sander (2016), Monthly trends of methane emissions in Los Angeles from 2011 to 2015 inferred by CLARS-FTS observations, Atmos. Chem. Phys. Discuss., (April), 1–29, doi:10.5194/acp-2016-232.

Wunch et al (2009) used CO2 instead of CO as the basis to estimate CH4. Also,Wunch et al (2009) pointed out the possible underestimation of CH4 if it was computed from CO emissions, given their differing diurnal profiles (CO emissions primarily influenced by traffic, which was believed to be a stronger daytime source than methane). This new discussion manuscript does not address these issues. The authors should clarify how potential differences in the diurnal profiles of CO, CH4, and C2H6 could affect the emissions estimates calculated with Equation 4.

Subsequent work has better agreed with the (lower) emissions estimates calculated using CO using aircraft and other remote sensing techniques. A sentence to this effect has been added to the Methods section:

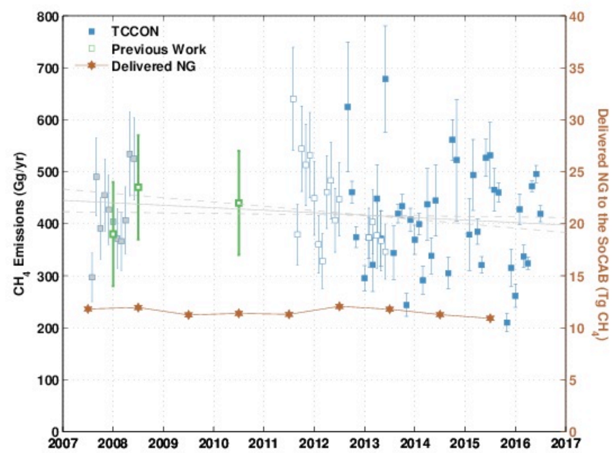
Wunch2009 suggested that using CO instead of CO2 to compute emissions may underestimate the emissions due to different diurnal emissions patterns, but subsequent studies have shown better agreement with the CH4 emissions estimates computed using its relationship with CO [Wennberg2012,Peischl2013,Wong2016].

The authors should emphasize the importance to their analysis of the changing C2:C1 ratio in pipeline gas. This trend appears to serve as tracer of opportunity, a unique fingerprint that allows attribution of the total observed C1 signal to infrastructure associated with handling, storage, delivery and use of pipeline quality natural gas. This is done indirectly in line 220, but the scientific novelty and utility of the trend deserves greater attention.

Agreed!

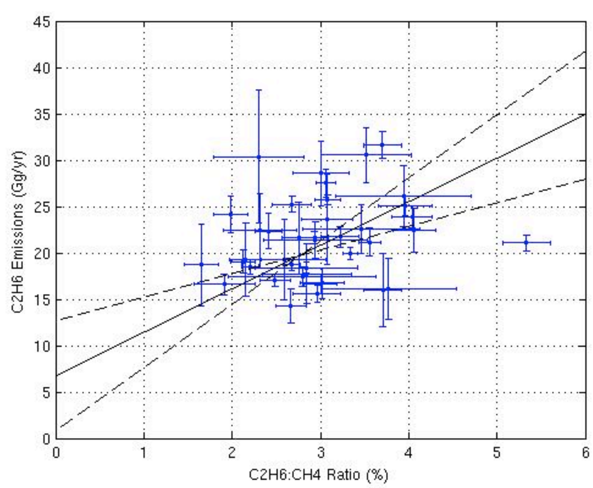
The manuscript's impact would be improved if the authors could provide a more complete picture about the contribution of specific source types to the observed C1 and C2 trends. Having partitioned the fraction of total methane signal due to pipeline gas (possible due to its increasing ethane content), can the authors further delve into the individual methane and ethane trends and provide a conceptual model that explains the recent trends or patterns in monthly/annual C1 and C2 emissions. [It would seem the C2 emissions might be reducible to a 2-source model (pipeline gas and associated gas/geologic seepage) with appropriate adjustment for vehicle emissions. Similarly, C1 emissions might be reducible to a 3-source model, by adding a generic third term for biogenic C1 sources.] At a minimum, the authors should clearly indicate whether the increasing C2:C1 ratio in pipeline gas is, by itself, sufficient to explain the potentially increasing C2 trend in Fig 3 and 6? Or can the balance of the C2 budget not explained by pipeline gas losses be explained: for example, given likely associated gas compositions, could the local oil/gas production to which Peischl attributed 32 Gg C1 also account for the excess C2 that is not explained by losses of pipeline quality gas? Alternatively, are other causes required?

There has been a very small decline in the methane emissions over the past 8 years, but no statistically significant change since 2012, when the C2:C1 ratios began increasing. We've created a new figure to show this, that makes use of two TCCON stations that were temporarily in the SoCAB in 2007-2008 and 2011-2013. Overlaid on this plot is the natural gas delivered to SoCAB customers with the y-axis scaled to match the left y-axis if 2% of the natural gas is lost as fugitive emissions.



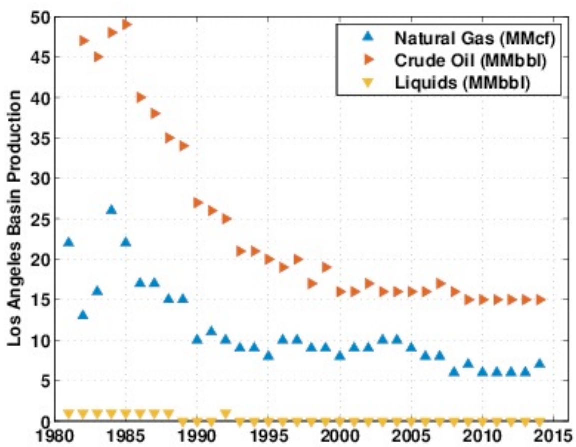
Assuming a constant methane emission over the 2012-2016 period, the C2:C1 ratio in the pipeline gas is sufficient to explain the increase in ethane since 2012: if we assume CH4 emissions are 413 Gg/yr, and roughly 240 Gg/yr from pipeline natural gas, we would infer C2H6 emissions of 11.6+/-4.4 Gg/yr in 2012-2013, 13.3+/-5.0 Gg/yr in 2013-2014, 15.0+/-5.7 Gg/yr in 2014-2015 using the increasing ethane to methane relationship. Adding this to the Peischl et al. 2010 estimate of C2H6 emissions from local oil and gas, vehicles, and the CARB "other" category (5.4+/-1.0 Gg/yr) results in 17.0+/-4.5 Gg/yr, 18.7+/-5.1 Gg/yr, and 20.4+/-5.7 Gg/yr for 2012-2013, 2013-2014, and 2014-2015, respectively. This falls well within the uncertainties of the ethane emissions estimates from the correlation with CO (19+/-4 Gg/yr 2012-2013; 21.4+/-4 Gg/yr 2013-2014; 23+/-3 Gg/yr 2014-2015).

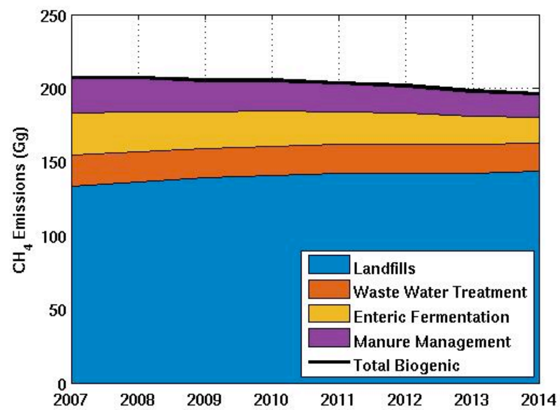
Attempts to extrapolate this relationship between ethane emissions and C2H6:CH4 ratio back to a regime in which the C2H6:CH4 ratio in the natural gas is zero (to get a sense of the magnitude of C2H6 in the SoCAB in the absence of natural gas C2H6 emissions) is not possible, due to the significant uncertainty on both the monthly C2H6:CH4 slopes and monthly C2H6 emissions. The y-intercept is 7+/-6 Gg/yr, implying that natural gas can explain anywhere from 1/3 to all of the C2H6 in the SoCAB atmosphere.



Once the C2 budget is determined, and knowing the C2:C1 ratio of pipeline gas, what can the authors say about the trend in C1 emissions due to losses of pipeline quality gas? It would be valuable if the authors could provide an assessment of whether the data indicates that downstream natural gas emissions in the region are changing.

We can say from our measurements that the methane emissions were roughly constant between 2007-2016 (changing by -5+/-4 Gg/yr), and no statistically significant decline is seen over the 2012-2016 period. According to the EIA, the oil and gas production in the Los Angeles Basin (somewhat larger than the SoCAB) has remained relatively constant over this period (left plot below). Biogenic emissions from the CARB statewide inventory scaled to the SoCAB totaled about 207 Gg in 2007, declining by about 1 Gg/yr due to a 2.8 Gg/yr livestock population change and a partially compensating increase in landfill emissions (right plot below). Thus the small decline we see in atmospheric CH4 emissions (-5+/-4 Gg/yr) might be partially attributable to the decline in biogenics, but the uncertainties are too small to be confident. The downstream natural gas emissions do not appear to be changing significantly.





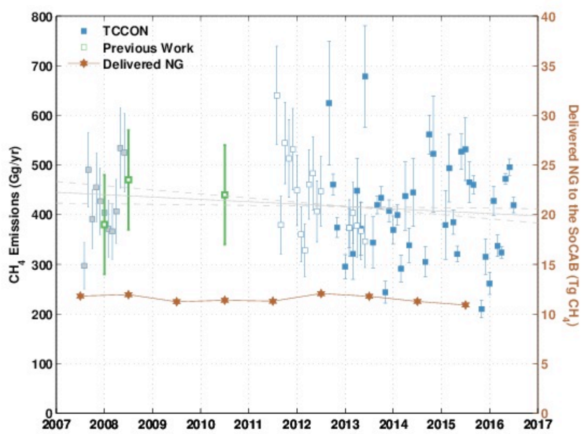
It is not clear from the text at line 225 and the reference cited how the authors derive the mass of C1 delivered by SoCalGas to customers within the SoCAB. It is also unclear why sales data going back to 2003 are relevant at this point of the discussion focused on the regional methane budget in 2015 (it would be more relevant – indeed desirable – to show historical gas deliveries in Figure 4). Southern California Gas' annual report (Sempra Energy 2015 Financial Report) reported annual volumes of gas sold in 2013, 2014 and 2015 of 999, 944 and 925 bcf (average 960 bcf, or 17.5 Tg assuming a methane content in gas of 95%). The authors should explain how they partition the SoCalGas' systemwide sales to isolate the customers solely within the SoCAB. Because not all of the gas sold by SoCalGas is consumed within the SoCAB and may or may not be transported through the SoCAB, the authors should report multiple metrics for the loss of pipeline quality gas that is sold or transported across the basin. One metric would be % of methane delivered that is emitted, and the other is the emissions as a percent of methane throughput in the SoCalGas system. The latter yields a loss rate for pipeline gas of 1.4% of potential throughput (242Gg/17.5Tg). The comparison to Wennberg et al's 2% loss rate should be done with caution, ensuring that the quantities in the numerator and denominator are apples-to-apples between this work and the previous work (it seems

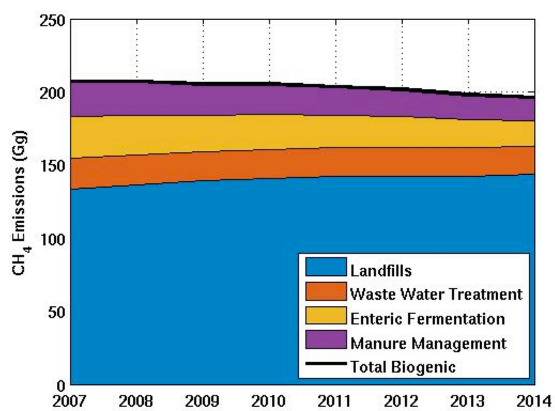
the 2% in Wennberg would most appropriately be compared to 1.4%, as calculated above).

Agree this was unclear. SoCalGas have now published their 2015 delivery numbers, so historical data are no longer necessary. Here is the reworked paragraph:

Since the average total methane emissions in the SoCAB since 2007 have been roughly constant at 413+/-86 Gg yr^{-1}, the ~58\% attributable to the natural gas infrastructure is 240+/-78 Gg yr^{-1}. In 2015, the SoCalGas total throughput was 2559 MMcf day^{-1}, or 18 Tg CH4 total. We remove 3 Tg CH_4 from wholesales, and 0.2 Tg CH_4 from company use and ``lost and unaccounted for'' (LUAF) gas, giving 14.7 Tg CH4 delivered by SoCalGas. This suggests 1.6+/-0.5% losses as fugitive emissions from the total delivered. (However, only 74\% of the population served by SoCalGas lives in the SoCAB, and thus the fraction of the losses as fugitive emissions would represent a larger fraction of the delivered gas to SoCAB customers [Wennberg2012].)

Figure 4. The manuscript would be improved if the hydrocarbon production data provided was specific to the SoCAB rather than statewide (these are publicly available from state agencies). Additionally, since hydrocarbon production is only a small contributor to C1 and C2 emissions in the SoCAB, this figure would be much more useful if it presented publicly available activity trends for other chief sources – in particular, I would suggest SoCalGas' natural gas sales and livestock populations. Recent CH4 emissions data or landfills and waste water treatment plants may also be available through the US EPA Greenhouse Gas Reporting program or California state equivalents.





We have now included Los Angeles Basin production instead of statewide data, which simplifies this analysis somewhat. Furthermore, the left plot above shows the delivered natural gas to the SoCAB (right axis), which is about 11 Tg/year, and the roughly constant CH4 emissions we compute since 2007 (left axis) from our atmospheric measurements. If we assume 2% fugitive emissions, this delivered natural gas represents about 220 Gg/year. The right plot is the emissions from the CARB emissions database for California landfills and wastewater (scaled by SoCAB population relative to California), and enteric fermentation and manure management (scaled to the cattle and calve population in the SoCAB counties relative to California). As described earlier, these biogenics totaled about 207 Gg in 2007 and change only slightly in time. We have therefore added an inventory table that uses our measurements, the 2010 Peischl et al. inventory for biogenics, local oil and gas and vehicles, and included our pipeline natural gas emissions. We compare the sum of the inventory to our atmospheric estimates and the results agree within uncertainties.

The richest findings seem to derive from the more recent and denser Caltech FTS measurements, with the JPL MkIV FTS data providing corroboration and further insight about historical trends. The manuscript's flow and clarity might be improved with some reorganization of the results and discussion or more explicit delineation of how the two data sets are used to support the conclusions reached.

We reworked the paper with this in mind.

The results relating to Aliso Canyon are interesting and important, but are not central to the paper's main findings. I would recommend moving the Aliso Canyon discussion into a separate subsection.

Done.

Abstract Line 9. The introduction of "Our methane emissions record" here is confusing since line 4 refers to a record dating back to the 1980s.

Corrected and reorganized the abstract.

Abstract Lines 10-15. This wording might be misconstrued to imply that the source of the excess methane is the gas storage facility. In fact the gas storage facility is only mentioned since it is a reliable source of C2:C1 ratios. But the authors have a secondary data source (delivered gas) that yields a statistically indistinguishable trend line in Fig. 5. The authors should revise the language to indicate the comparison is between atmospheric measurements and measured C2:C1 of gas delivered and stored in the region. Additionally, the authors should more explicitly indicate the scope of natural gas infrastructure implicated in the final sentence – to indicate it includes gas delivery infrastructure including pipeline leaks (transmission and distribution), compression and storage facilities, and post-meter losses among others.

Reworked abstract.

Line 179. It was unclear how the statement about ethane to acetylene ratios followed from statements about C2:CO and acetylene:CO; please elaborate on the significance.

Updated text:

There are three main sources of ethane emissions in the SoCAB: vehicle exhaust, the natural gas system, and oil and gas exploration and extraction. Of these sources, only vehicle exhaust is not a significant source of CH4. To distinguish between vehicle exhaust and fossil fuel sources, we use our coincident measurements of carbon monoxide, which tracks sources of incomplete combustion (including mobile sources), and acetylene (C2H2), whose emissions more directly track vehicle exhaust [Kirchstetter1996,Warneke2012,Crounse2009]. The ratio of ethane to carbon monoxide in the SoCAB declined rapidly until the mid-1990s, and then slowly and steadily increased. The ratio of acetylene to carbon monoxide remained relatively constant throughout the time period, and thus the ethane to acetylene ratios follow the same trend as ethane to carbon monoxide. This implies that vehicle emissions are not driving the changes in ethane emissions. This is consistent with the Warneke2012 analysis, which showed an increase in ethane relative to acetylene after 1995, which they attributed to natural gas use and production.

Line 226. The statement attributing 242 Gg/yr C1 to natural gas infrastructure should be linked back to the prior paragraph's finding that 54% of total excess was due to natural gas (e.g. "242 Gg/yr, equal to 54% of the SoCab total. . .".

Done.

Lines 248-255. The specific value used for GWP100 should be stated (e.g., 25, 28, or 34). The choice of 100-yr GWP in this paragraph does not account for the greater short-term climate impacts of CH4. The authors should consider reporting a 20-yr CO2e value in addition to the 100-yr value. The reference to climate impact in the last sentence needs to explicitly distinguish short- and long-term impacts; if only 100-yr GWP comparisons are made, then the sentence should be clarified to refer to "longterm climate impact. . ."

Done.

Figure 1. The very rapid rise in C2 mole fraction in the most recent JPL MkIV FTS measurements should be explained (panel 3). Is this trend due to the increased C2:C1 ratio, the Aliso Canyon blowout or both? Should the C2 rise be accompanied by changes in C1?

Those six high C2 points are on a single day (November 10, 2015), and are due to the Aliso Canyon blowout plume having been advected over the line of sight of the MkIV instrument. The C2 rise is accompanied by a smaller (2.5%) increase in C1, which is difficult to see in the raw CH4 data due to natural variability, but by plotting CH4 versus N2O (see slide 14 of: http://mark4sun.jpl.nasa.gov/report/MkIV_ethene_Toon.pdf), the CH4 increase becomes much clearer.

Line 141. The word "are" appears twice.

Removed.

Figure 1. The black Mauna Loa data points are significantly obscured by the CO and C2 data points.

Revised figure.

Fig 3. The error bars are hard to make out and the symbol for the Peischl et al is not evident.

Figure clarified.