Referee comments in black. *My responses in green italics.*

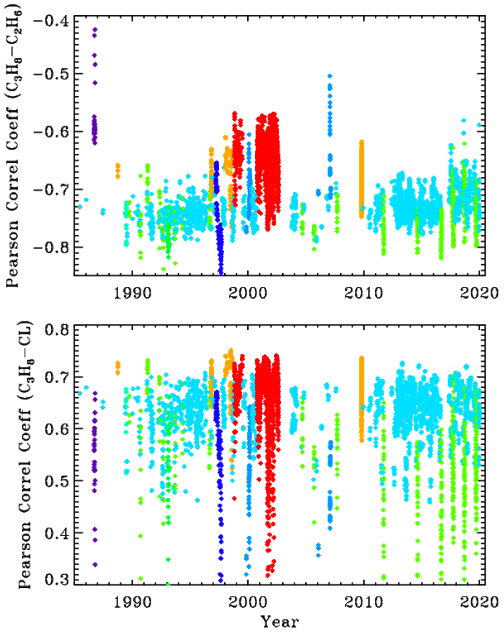
Anonymous Referee #1

The paper decribes the first spectroscopic measurements of atmospheric propane (C3H8) using the solar absorption spectrometry. The paper is very well written, very interesting, and should be published. ACP is the right journal. *Thank you!*

The paper states that 5000 ground-based spectra have been recorded with the Mark4 at 1200 days, and 12 different sites (lines 59-60). Figure 2 gives a result for the average of all 5000 spectral fits. I assume that these 5000 spectra have been recorded at a variety of many different solar zenith angles. Also, I assume the figure gives the average of the retrievals for spectra recorded at very different altitudes, from the ground up to 2.5 km altitude. I do not see that its possible to average the results of these very different retrievals. This might be possible, but requires an explanation. Or, is it possible to show the result for one spectral fit?

We agree that due to the non-linearity of transmittance with airmass, the effect of averaging spectral fits taken at different SZA or altitudes does not give the same result as would be obtained at the average SZA and average altitude, especially for strong absorption lines. So it would not be valid to fit an average spectrum. But since we fitted individual spectra and then averaged, the resulting spectral fit provides a valid representation of the residuals of the overall dataset, with the advantages of reducing the noise level and some systematic errors, e.g., due to atmospheric temperature errors. If we were to use a single spectrum fit, which site, zenith angle, and propane amount would we choose? Whatever spectrum I choose, it would be representative of only a small fraction the dataset.

The paper discusses in several Figures the correlation between C3H8 and C2H6. When I look at Figure 2, it seems that both trace gases absorb in the same spectral region. The paper mentions ... litte spectroscopic "cross talk" between these two gases (line 109) but it might be useful to discuss this a bit more quantitatively, if possible. I accept that the absorption features of both trace gases are very different, but a small discussion would be helpful.



The following new material is now included in the paper as Appendix A. We compute Pearson Correlation Coefficients (PCC) from the a posteriori covariance matrix for each of the 5000 spectral fits. The upper panel (left) shows the PCC between retrieved C_3H_8 and C_2H_6 for the 5000 spectra. Points are plotted versus year with the same site-altitude-dependent coloring as in Fig.3. The PCC between C_3H_8 and C_2H_6 averages about -0.7, which means that they are fairly strongly anti-correlated. This is due to their overlapping absorption features at 2967.5 cm^{-1}. So as retrieved C_2H_6 increases, C_3H_8 will decrease, and vice versa. The PCCs are closer to zero for the high-altitude sites (red & orange), presumably due to the reduced pressure broadening and decreased H_2O absorption causing the C_2H_2 and C_2H_6 absorption spectra to become more distinct. This anti-correlation could be reduced by use of a wider window to introduce additional C_2H_6 features that don't overlap the C_3H_8 Q-branch, but this would likely also encompass large residuals without adding any C_3H_8 information.

Also shown (lower panel) is the correlation between C_3H_8 and the fitted Continuum Level (CL). These have a correlation of about +0.65 at low SZA, decreasing at higher SZA. So the more C_3H_8 that is retrieved, the higher the continuum level has to be to match the measured spectrum, due to the fact that the C_3H_8 absorption spectrum has a broad continuum-like component. The PCCs between C_3H_8 and the other retrieved parameters (e.g., H_2O, HDO, CH_4, Continuum Tilt, Frequency Shifts) were all much closer to zero than with C_2H_6 and CL.

The high PCC between C_3H_8 and C_2H_6 doesn't necessarily imply a large uncertainty in the C_3H_8. It just means that the large component of the C_2H_6 uncertainty gets projected onto the C_3H_8. Ditto for the CL. But provided the C_2H_6 and CL are well retrieved, their effect on the C_3H_8 will not dominate.

The comparison between in-situ and total column data is difficult. However, using the assumed a-priori profile used, together with the scaling factor for the retrievals, it is possible to get a rough idea on the surface concentration. This would be useful for the comparison. It could show a good agreement, or that the a-priori assumption is wrong (assumed that the spectroscopy is correct).

Agreed. But the retrieval returns just one piece of information on C_3H_8. So the deduced surface concentration would be highly dependent on the assumed a priori, which is highly uncertain under conditions of enhanced C_3H_8.

Minor comments: The absorption of C3H8 is quite weak, and the retrieval has been performed very carefully and at its limits. It would be good to have a small discussion about the possibility to measure C3H8 at other NDACC sites. Does it make sense that other groups look at C3H8 in their spectra?

Yes, NDACC sites in polluted locations should look for C_3H_8. For sites in clean locations, however, the spectroscopy needs further improvement to be able to obtain useful measurements under background C3H8 levels. The strong H2O line at 2966.0 cm^{-1} is particularly troublesome in driving up the fitting residuals. We have modified the summary section to further emphasize this point.

Figures 3, 4 and 6 contain a lot of information, but since data from different sites are in one Figure, the reader might get the impression that the overall scatter is extremely high, and that the measurements are useless. I suggest to start with Figure 10, which gives a clear and nice indication of the measurements and their limits. The other results might also be presented differently, may be showing the results for the different sites individually.

Figure 10 contains just the JPL data, one of 12 observation sites. And it plots Xgas values. I continue to prefer to start with an overview of the whole dataset, show the raw column data, then explain how to derive Xgas values from the columns, then look at individual sites. Yes, some readers might initially think that the measurements are useless, but those that persevere will discover that C3H8 is highly variable.

For Figure 11, left it might make sense to plot annual averages instead of the individual data. This holds also for several other Figures.

Not sure what is gained by plotting annual averages? Sure, it will reduce clutter in the SGP panels, but information in the variability is lost.

Line 37: ... essentially zero at ... What is essentially zero? Please give a concentration, or upper limit.

Replaced "essentially zero" with "below 50 ppt".

Line 143-146: As far as I understand, its written: At lower altitudes the uncertainties are quite high. In the next sentence its written: This allows to detect C3H8 under polluted conditions. I dont understand these two sentences, for me this is contradictory.

Good point. Sentence has been reworded as " But the C3H8 increases far more, allowing C_3H_8 *to be detected at these low-altitude sites under polluted conditions, despite the poorer absolute uncertainties."*

Line 148: 'use' should be 'us' *Fixed.*

Line 203-209: I dont see why it is important to discuss CH4. For me this chapter could be deleted.

I agree that CH4 is not the main thrust of this paper. But CH4 is the main constituent of NG and is well-measured by the MkIV, so it would seem remiss, or even evasive, not to show some MkIV CH4 data and explain their limitations toward diagnosing the causes of the C3H8 enhancement.

Line 266: The estimation given in this chapter is interesting, but I have the feeling that the 4%, given in line 266 is not an assumption, but a result, giving meaningful data. This chapter might be rewritten.

Yes, you are right. I have re-written this paragraph. I have also realized that the Permian Basin Iol and NG field is larger than I originally estimated. This is also corrected.

Line 278: I would suggest to include the averaging kernel figures in the main text.

Done. A new section was created "3.1 Averaging Kernels".