

In this manuscript, the authors investigate the overlap of TES peroxyacyl nitrate (PAN) detections (defined as $\text{DOF} > 0.6$) with HMS smoke extent and TES CO retrievals in the western United States. The authors 1) quantify the fraction of “enhanced” TES retrievals ($\text{DOF} > 0.6$) overlapping HMS smoke extent by month and year, 2) perform two case studies using FRAPPE, HYSPLIT, MODIS and CALIPSO data and 3) evaluate the ratio of TES tropospheric average CO and PAN enhancements.

Based on feedback from preliminary reviews, the authors have removed comparison of TES PAN measurements with model simulations from the manuscript but have not substantially improved the quantitative characterization of the retrieved product.

We respectfully disagree with the reviewer on this point. The re-submitted version included an additional figure (Figure 1) that demonstrates the limitations in the sensitivity of TES PAN measurements.

Additionally, there are editorial and typographical errors in the manuscript (e.g., Figure 6 top panel appears almost identical to Figure S3; seemingly erroneous in text figure references – Line 178).

We have corrected the caption for Figure 7. As discussed in response to Reviewer 1, there was a mix up in reference to “black” versus “red lines” from the earlier version. Figure S3 has not been changed as we intend it to be very similar to the top panel of Figure 7 (originally Figure 6). However, S3 shows additional trajectories than the version in the main manuscript.

Further characterization of TES PAN retrievals is important and the authors have worked towards that goal.

This comment seems in contrast to the one above from this reviewer, and we assume that it refers to the addition of Figure 1, which demonstrates the limitations in sensitivity of the TES PAN measurements. Perhaps the earlier comment was written before noticing Figure 1.

However, I recommend that the authors use language that more precisely conveys the uncertainty of the product. In the introduction, for example, the authors state that “Satellite measurements [i.e., TES] are essential to understand the seasonal cycle and interannual variations of PAN (L65).”

Here and similarly throughout, use of a strong word like “Essential” connotes long-standing maturity and widespread use. In this case, I would recommend using a “potential tool” instead of “essential”. While this is one specific example, edits should be made throughout the manuscript to address this concern.

We have changed the wording here as suggested and have re-read the manuscript to remove other similar instances, but the existing set of aircraft data is insufficient to identify and understand the seasonal cycle and interannual variations in PAN in the free troposphere. The one exception is the very recently collected AToM observations, and this mission has not yet ended. The specific wording has been adjusted to read:

“Given the limited set of long-term in situ measurements, satellite measurements are a potential tool that can be used to investigate the seasonal cycle and interannual variability of PAN in the troposphere along with which processes contribute to these features.”

The examples below indicate where the authors can add information to the manuscript from their already accomplished analyses that will be useful to the reader.

In addition to the specific edits listed below, we have substantially increased the information in Section 2.1 in response to some suggestions by Reviewer 1. These edits add quite a bit more information about the nature of the measurements.

Figure SI 4 seems to imply that there is no statistically significant difference between “In smoke” and “Not in smoke” retrievals. Is this expected? This needs to be addressed in the main text.

This is already addressed directly in the main text in Section 3.1, but we have added a few more sentences here. This result is not surprising because the HMS smoke polygons only indicate that there is smoke in the

column. They do not indicate whether the smoke is in the free troposphere (i.e. TES can detect it) or primarily in the boundary layer, where TES has weak sensitivity. When we restrict the TES data to only those retrievals that also have elevated CO in the free troposphere (TES 510hPa CO > 120 ppbv or TES 510hPa CO > 150 ppbv), we do see a difference between the “in smoke” and “not in smoke” retrievals.

One of the paper’s main findings is that ~15-32% of PAN detections (DOF > 0.6) overlap with HMS smoke extent. Is that a larger or smaller number than expected? Please report the percent of all attempted TES retrievals that overlap HMS smoke extent for context.

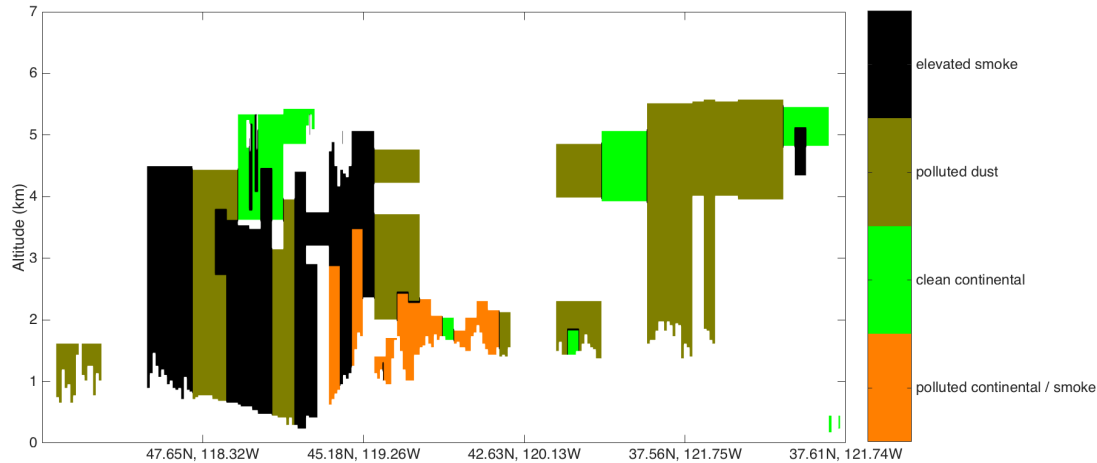
We have added this information to Section 3.1. This now reads: “Of all the retrievals attempted in July 2006 to July 2009, the percent associated with smoke is 18%. We expect a higher fraction of overlap in the subset of data with DOF > 0.6. This threshold value of DOF > 0.6 is consistent with a signal to noise ratio (SNR) greater than 1 (Payne et al. 2014), and this subset of data only reflects conditions with elevated PAN in the atmospheric column.”

The authors report that there is no statistical difference between PAN “detections” (DOF > 0.6) that do and do not overlap HMS smoke extent. Please discuss this finding in context of past airborne or mountain-top in situ studies: should we expect there to be a statistical difference in PAN concentrations in biomass burning influenced air and other polluted airmasses.

See our response above. HMS smoke polygons only indicate that there is smoke in the column. They do not indicate whether the smoke is in the free troposphere (i.e. TES can detect it) or primarily in the boundary layer, where TES has weak sensitivity. When we restrict the TES data to only those retrievals that also have elevated CO in the free troposphere (TES 510hPa CO > 120 ppbv or TES 510hPa CO > 150 ppbv), we do see a difference between the “in smoke” and “not in smoke” retrievals. We have added several additional references to point readers to aircraft and surface measurements of PAN enhancements in biomass burning plumes. However, we note that we use aircraft data in the paper already (Figure 9). Without an enhancement in PAN above the background, our calculations would not be meaningful. Elevated PAN is often observed in biomass burning plumes.

There is one case study involving CALIPSO data. The paper might benefit from a more statistically robust analysis of CALIPSO and TES data. The meridional offset of ~500 km between the two sensor tracks (Figure 6) could likely be overcome by the incorporation of reanalysis wind products and the potentially higher quality smoke information provided by CALIPSO.

It would be interesting to do a comparison between TES and CALIPSO data, but that is beyond the scope of this paper, and would need to be focused carefully. The point of Figure 6 (now Figure 7) is to show that the smoke was likely located in the lowest 5 km of the atmosphere. We also examined CALIPSO Aerosol data from an overpass located further west on 23 July 2007. There is polluted aerosol in the column during this overpass as well. The southern portions of this overpass are located west of the active fires on this day. Similar to the image that we show in Figure 6 (now Figure 7), smoke or polluted smoke is identified from the surface to 4-5 km. The swath shown in Figure 6 overlaps an HMS smoke polygon that extends from the WA/ID border to Minnesota. We also note that the trajectories displayed in Figure 6 are based on reanalysis wind products.



CALIPSO aerosol subtype observed on 23 July 2007, 10:23:44.5. CALIPSO Science Team (2016), CALIPSO/CALIOP Level 2, Vertical Feature Mask Data, version 4.10, Hampton, VA, USA: NASA Atmospheric Science Data Center (ASDC), Accessed by Emily V. Fischer at doi: 10.5067/CALIOP/CALIPSO/LID_L2_VFM-Standard-V4-10

“The TES PAN retrievals shown here were processed using a prototype algorithm for the area and time periods of interest.” Please provide more details regarding differences between this prototype and the operational retrievals. What is the source of a priori profiles in the retrieval? What is the tropospheric average PAN concentration in the a priori profiles? The prior profile shown in Figure panel 2d shows PAN concentrations between 300 ppt and 400 ppt – if so, please justify use of an assumed “background” concentration of 100 or 200 ppt in the PAN/CO enhancement analysis in Section 3.3.

The prototype algorithm used here is effectively identical to the algorithm that has been implemented in the v7 routine Level 2 processing. We have now added further information to this effect in the text. Background PAN or CO refers to that not strongly impacted by smoke. It does not refer to the a priori.

“For footprints where the spectra show strong evidence of this silicate feature in the surface emissivity (this can occur over rocky or sandy surfaces) TES PAN retrievals are not attempted.” What fraction of retrievals is discarded by this requirement? Does dust aerosol have a similar silicate absorption or emission feature that should be considered in the retrievals?

In early testing of the algorithm, we had found that desert and rocky surfaces would tend to show high initial chi-squared values for the PAN retrieval, with residual features that could only be brought within the noise by fitting strongly negative PAN VMR values. Further explanation of this issue, with a figure, is shown in Payne et al. (2014). Aside from negative values being unphysical, the current algorithm retrieves in terms of $\ln(\text{vmr})$, so negative PAN values are not possible to retrieve within the current framework. In general, we had found that cases with high initial chi-squared values would tend to fail, and we had chosen to set a threshold (initial chi-squared > 3.0) above which retrievals are not attempted. Of course, surface emissivity features are not the only reason why there might be a high initial chi-squared value. Another reason might be poor fits to interfering species, such as water vapor. In the current version of the algorithm, we do not attempt to explicitly track all the different reasons for failure of quality control, but instead have implemented a master quality flag. We have added the following text to address the reviewer’s question about the fraction of retrievals discarded:

“Of the 28149 TES footprints processed for this work that fell over land, 3608 of them failed quality control. Concentrated regions of failed quality control show up as white patches in Figure 2(b). These regions are largely desert or mountainous regions.”

The reviewer raises an interesting point about dust aerosol. There could indeed be silicate features in dust aerosol. A number of groups have looked at dust signatures in spaceborne thermal infrared radiance measurements and at their impact on other retrieval products. If the dust absorption were strong enough, this could be another reason why the TES PAN retrieval might not be attempted due to a high initial chi-squared value. If the dust absorption were sufficiently weak, this would cause the TES PAN retrieval to be biased low. A TES dust flag or dust product does not currently exist, and a rigorous assessment of the impact of dust aerosol on the TES trace gas products is outside the scope of this study, but would be worthy of consideration for future work. We have added two sentences in the manuscript on the possibility of impact from dust aerosol, with example references, and thank the reviewer for raising this point.

L118 – What is “extremely elevated”? Please quantify.

We have removed the words “extremely elevated”, and this now reads:

“Mean PAN observed by the C-130 below 3 km during the field campaign was 481 pptv (Zaragoza et al., 2017). This particular day (29 July) was one of the four days identified by Zaragoza et al. (2017) with the highest surface PAN mixing ratios observed at the Boulder Atmospheric Observatory.”

L130 – “We only include data with DOFS > 0.6 to ensure that the retrievals are dominated by real observed information.” “Dominated” is a rather strong word to describe DOFs > 0.6. The meaning of “real observed information” is unclear.

In response to the other reviewer, we have replaced this sentence with more specific information. This now reads:

“We only include data with DOFS > 0.6. More specifically, this threshold value of DOF > 0.6 was chosen to be consistent with a signal to noise ratio (SNR) greater than 1 (Payne et al., 2014), and this criteria has been used in all the papers that have presented TES PAN data thus far (Zhu et al., 2015; Payne et al., 2016; Jiang et al., 2016; Zhu et al., 2017).”

L132 - “This conservative choice means that we are primarily basing our analysis on retrievals with high PAN” What is the mean and standard deviation of retrieved PAN concentrations?

We have added the following sentence: “The mean (standard deviation) of the retrieved tropospheric average PAN mixing ratios for DOFS > 0.6 for the region shown in the figures presented here (125° W - 70° W, 30° N - 50° N and 130° W - 65° W, 50° N - 70° N) is 0.551 (0.925) ppbv.” However, this can be seen in the supplemental figures.

L172 – 193 – The purpose of this paragraph is not clear to me. Furthermore the paper evidence is not all that convincing as the expected relationships between smoke and TES PAN detections is not consistent or as expected. I did not find Figures 3 or 5 to be particularly helpful either.

The point of this paragraph is to show the spatial distribution of which TES retrievals overlap HMS smoke plumes each month. We have decided to keep all these figures in the main body of the manuscript. Figure 4 (now Figure 5) shows the distribution of tropospheric average PAN and CO as measured by TES within smoke plumes over the U.S. Figure 5 shows which day of the month overlaps a smoke plume. These figures are essential for the reader to understand the data, and the other reviewer did not make any negative comments about these figures.

L194-210 + Figure 4 – The data in this paragraph and figure can be used to compute a PAN:CO ratio that is less dependent on assumptions of background contributions. Based on a cursory visual analysis of Figure 4, a value of 0.3% PAN:CO appears reasonable. Figure 4 – The data that is “not in smoke” should be separated similarly to the data that is “in smoke.” Is the relationship between CO and PAN similar “not in smoke” retrievals similar to their relationship “in smoke” (see above comment)? Should that result be expected based on previous comparisons of anthropogenic (not in smoke enhancement) and biomass burning influenced (in smoke) air?

Sure, this would be another approach, albeit a bit coarse. This quick visual approach does yield a value that agrees with many of our samples (see Figure 8 histogram). However, giving one value like this would not show the range of PAN enhancements that are observed. It does not make sense to present a PAN enhancement ratio relative to CO in non-smoke impacted samples. In these samples, the PAN and CO could have different sources. We do not know if the other samples are anthropogenically influenced. The PAN could also have been produced by lightning NO_x. Or the PAN and CO could be at different levels of the atmosphere. This is why we have been very conservative, presenting enhancement ratios only using the small subset of data where 510 hPa CO is great than 150 ppbv.

Editorial comments: Below is a non-exhaustive list of editorial suggestions or errors. I recommend that the authors thoroughly proofread before re-submission.

L1 – Re-define PAN as peroxyacyl nitrate in the body of the manuscript.

PAN is commonly known by its misnomer peroxyacetyl nitrate. This is a very minor point, and this name is used in many other manuscripts. However, we have changed the name as requested.

L140 – “infrared” [imagery]. L174 “NOAA HMA smoke plume”

Suggested change has been made.

L178 – “Supplemental Figure 1” – Perhaps intended for Supplemental Figure 2

Yes, this was also noted by Reviewer 1, and it has been corrected.

Figure S3 and Figure 6 top panel appear to be identical.

Figure S3 has not been changed as we intend it to be very similar to the top panel of Figure 7 (originally Figure 6). However, S3 shows additional trajectories than the version in the main manuscript. S3 looks a bit too cluttered for the main body of the paper, but it provides more information.