

We thank the reviewers for taking the time to review our paper and for their helpful comments, which have helped us in strengthening our paper. Below we address each comment in turn, and describe how it has been addressed in our revised manuscript. Reviewer comments are in bold, and our responses in normal text.

### **Reviewer #1**

**This work investigates the impact of using updated spectroscopic data on the analysis of TES spectra (as a part of the changes introduced in TES retrieval products version 6 over version 5). Aircraft observations from all five HIPPO (HIAPER Pole-to-Pole Observations) campaigns of the Carbon Cycle and Greenhouse Gases Study have been used for the evaluation of the induced changes. It should be noted that the new version 6 also incorporates an update of a-priori trace gas profiles, which to some degree reduces the conciseness of the present study (however, it is demonstrated that the induced changes are of secondary importance).**

**Altogether, this is a solid piece of work and I recommend publication in AMT. I would like to suggest consideration of the following minor corrections:**

**It is not clear to me whether surface temperature ( $T_{\text{surf}}$ ) is a retrieved quantity or not. The state vector for the joint estimate contains  $T_{\text{surf}}$ , whereas on the bottom of page 6, it is claimed that "The raw retrieved  $\text{CH}_4$  profile can have a significant positive bias due to errors in unretrieved model parameters such as ... surface temperature ... ."**

Surface temperature is indeed a retrieved quantity, along with the vertical profiles of  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ ,  $\text{H}_2\text{O}$ , and  $\text{HDO}$ , as well as the effective cloud pressure and optical depth, as shown in Equation 2 and discussed on Page 10065, Lines 6-8. The reference to surface temperature and clouds as unretrieved parameters on Page 10066, Lines 15-16 is thus incorrect, and we thank the reviewer for noticing the error so that we could correct it. We have removed this latter reference from our revised manuscript, which now reads (see Page 6, Lines 21-23):

The raw retrieved  $\text{CH}_4$  profile from TES ( $\hat{\mathbf{x}}_{\text{CH}_4}$ ) can have a significant positive bias due to errors in unretrieved model parameters (**b**) such as the temperature profile, surface emissivity, and spectroscopic parameters.

**Section 4.2 describes the application of the TES observation operator for HIPPO profiles. The method for extrapolation beyond the ceiling altitude is outlined. As section 4.1 states the lower bound of HIPPO profiles might be located at a level of up to 650 mbar, so the method applied for extrapolation downwards should also be provided for sake of completeness.**

We agree, and thank the reviewer for noticing the oversight in our original document. Given the well-mixed nature of CH<sub>4</sub> in the troposphere, especially over the remote ocean areas of interest in this study, we extrapolated the HIPPO profiles downwards assuming that the CH<sub>4</sub> concentration is the same as that at the lowest altitude measured, so that the HIPPO profile is constant with height below the lowest altitude. We have added this to our revised manuscript (see Section 4.2, Page 11 Lines 26-29 and Page 12 Line 1), which now reads:

To address this, we first interpolate each HIPPO profile (expressed as the natural logarithm of the mixing ratio) to the 67 levels of the TES pressure grid, extrapolating above the GV ceiling using the shape of the TES a priori profile (i.e., scaling the TES a priori to match the top of the HIPPO profile) and extrapolating below the lowest altitude measured by assuming that the profile is constant below this height.

**The caption of table 3 should contain the information that the tabellated values are in ppb**

We agree, and have made the requested change (see Page 34).

## **Reviewer #2**

**This article evaluates differences in a new processing version of TES CH<sub>4</sub> retrievals that employ an updated set of spectroscopic parameters for the molecule. It uses a reliable external reference data set (in situ aircraft measurements from HIPPO) that provides a good, independent measure of whether the new spectroscopic parameters improved results or made things worse.**

**The paper is well organized, and the English/grammar usage is excellent. The study is relatively thorough, and the conclusions are sound. I would recommend publication, subject to the following two technical corrections:**

**Page 10060, line 16: the acronym DOFS is used without definition**

We have corrected this in Page 2, Lines 1-3 of our revised manuscript, which now reads:

For retrievals with less than 1.6 degrees of freedom for signal (DOFS), the bias is reduced from 26.8 ppbv to 4.8 ppbv. We also find that updates to the spectroscopic parameters for N<sub>2</sub>O reduce the errors in the retrieved N<sub>2</sub>O profile.

**Page 10065, line 9: the retrieval grid vector,  $z$ , is defined in Eq. 2, not Eq. 1**

This has been corrected on Page 6, Line 3 of the revised manuscript

**My one area of concern with this work (for which I wouldn't necessarily suggest changes in the article) is the N<sub>2</sub>O correction. The authors do address the accuracy of their assumptions used as the basis of their correction (the N<sub>2</sub>O a priori). For measurements with at least 1.6 DOFS, the difference between the assumed TES N<sub>2</sub>O a priori and the HIPPO N<sub>2</sub>O measurements was on average <0.5 ppb, with a maximum of 5 ppb. For measurements with DOFS < 1.6 DOFS, the average discrepancy is about 1.5 ppb. They do not mention the maximum discrepancy in this data set. Deficiencies in the N<sub>2</sub>O a priori should not introduce significant error in the averaged results.**

**However, it seems to me that the N<sub>2</sub>O-based correction appears to work because the spectroscopic parameters employed for N<sub>2</sub>O (from HITRAN 2000) are "bad in a similar way" to the CH<sub>4</sub> spectroscopic parameters. When they switch to updated parameters for N<sub>2</sub>O, the retrieved N<sub>2</sub>O results are closer to expectations and the N<sub>2</sub>O-based correction no longer works properly. This is not an ideal situation for applying a correction. Ideally, deviations in retrieved N<sub>2</sub>O from expectations would be mostly from some inherent limitation in the retrieval process that would yield similar systematic errors in the CH<sub>4</sub> retrievals. Relying (at least partially) on the nature of deficiencies in the N<sub>2</sub>O spectroscopic parameters to correct CH<sub>4</sub> retrievals is a bit of a dangerous game, but it appears to improve the results and so would seem to be defensible.**

We do agree with the reviewer that the N<sub>2</sub>O-based correction is not ideal, as it does depend on the errors in CH<sub>4</sub> and N<sub>2</sub>O being similar, and while this is likely true for errors from surface emissivity, temperature profile, etc., there is no reason in principle that errors in the spectroscopic parameters have to be similar. Our use of the N<sub>2</sub>O-correction is thus based on the empirical evidence that it does improve the performance of the retrievals when the N<sub>2</sub>O spectroscopic parameters are taken from HITRAN 2000. However, as the reviewer notes, the new HITRAN 2008 N<sub>2</sub>O parameters do result in improved retrieved N<sub>2</sub>O profiles, but that means the N<sub>2</sub>O parameters are no longer "bad in a similar way" to the CH<sub>4</sub> parameters, causing problems with the correction procedure as noted on Page 10080, Lines 23-26.

**It would suggest to me that there likely remain errors in the CH<sub>4</sub> spectroscopic parameters. With the new CH<sub>4</sub> spectroscopic parameters, the agreement with HIPPO results improves, even in the absence of the N<sub>2</sub>O-based correction. I agree with the authors' assessment that the new set of CH<sub>4</sub> parameters represents an improvement. However, I suspect that further improvements are required in the parameters. There is a newer line list than was considered here (HITRAN 2012). It would be beyond the scope of this paper but is probably worth looking at. Based on my experiences with CH<sub>4</sub> in HITRAN 2012, I am not convinced it holds the answer, but I have not looked in detail at the frequency region employed in the CH<sub>4</sub> retrievals for this study.**

We agree with the reviewer that there are remaining errors in the CH<sub>4</sub> spectroscopy in this region and that further improvement is required. We have added the following sentence to Section 5.2 to make this explicit (see Page 15 Lines 1-3):

However, the remaining bias in the uncorrected CH<sub>4</sub> profiles from TES Version 6 suggest that further improvement of the CH<sub>4</sub> spectroscopic parameters is needed to remove the necessity of the N<sub>2</sub>O-based correction.

We also note that we are currently investigating the impact of the HITRAN 2012 CH<sub>4</sub> parameters on our retrieval algorithm, but are not yet able to make any definitive conclusions.

**Interestingly, the changes in spectroscopic parameters for N<sub>2</sub>O between HITRAN 2000 and HITRAN 2008 may hold some clue as to the sort of refinements necessary for CH<sub>4</sub>, if the HITRAN 2000 N<sub>2</sub>O parameters are “bad in a similar way” to the CH<sub>4</sub> spectroscopic parameters.**

This is an intriguing suggestion, and we will attempt to investigate this possibility in future work. We thank the reviewer for the suggestion.

**The residuals near 1310 cm<sup>-1</sup> suggest there might be something missing in the calculated spectrum. There is HNO<sub>3</sub> in that region, but this is not mentioned in the text. Does it not contribute significantly in your measurements?**

The major residual features near 1310 cm<sup>-1</sup> for TES Version 6 are associated with strong CH<sub>4</sub> and H<sub>2</sub>O lines (see Figure R1 below). The sharp negative residual at ~1311.4 cm<sup>-1</sup> is associated with a strong CH<sub>4</sub> line, the wide negative residual features at ~1312.6 cm<sup>-1</sup> and ~1314.8 cm<sup>-1</sup> are associated with strong H<sub>2</sub>O lines, and the negative residual at ~1316.8 cm<sup>-1</sup> is associated with both a H<sub>2</sub>O and CH<sub>4</sub> line. Thus we think these remaining residual features are more likely due to inconsistencies in the H<sub>2</sub>O and CH<sub>4</sub> spectroscopy rather than missing species. We have modified Page 13, Lines 11-15 to reflect this discussion:

The change to V006 has mixed results: while the spectroscopic parameters used in TES Version 6 do remove several of the strong residual features between 1190 and 1280 cm<sup>-1</sup> (especially the strong residuals near 1245-1247 cm<sup>-1</sup>), the spectral regions above 1290 cm<sup>-1</sup> show stronger residual spikes that are generally associated with strong CH<sub>4</sub> and H<sub>2</sub>O lines.

While HNO<sub>3</sub> is not included in the TES forward model in this region, the weak HNO<sub>3</sub> absorption and emission in this spectral region is a fairly broad feature with regularly spaced lines, and so this would be expected to impact the channels in this area similarly, rather than producing the observed negative residual spikes near strong CH<sub>4</sub> and H<sub>2</sub>O lines. The impacts of broad, weak spectral features, such as the HNO<sub>3</sub> absorption in this region, can be mitigated by the retrieval of the effective

cloud optical depth, which can account for the missing optical depth from these features as well as the optical depth of aerosols and clouds.

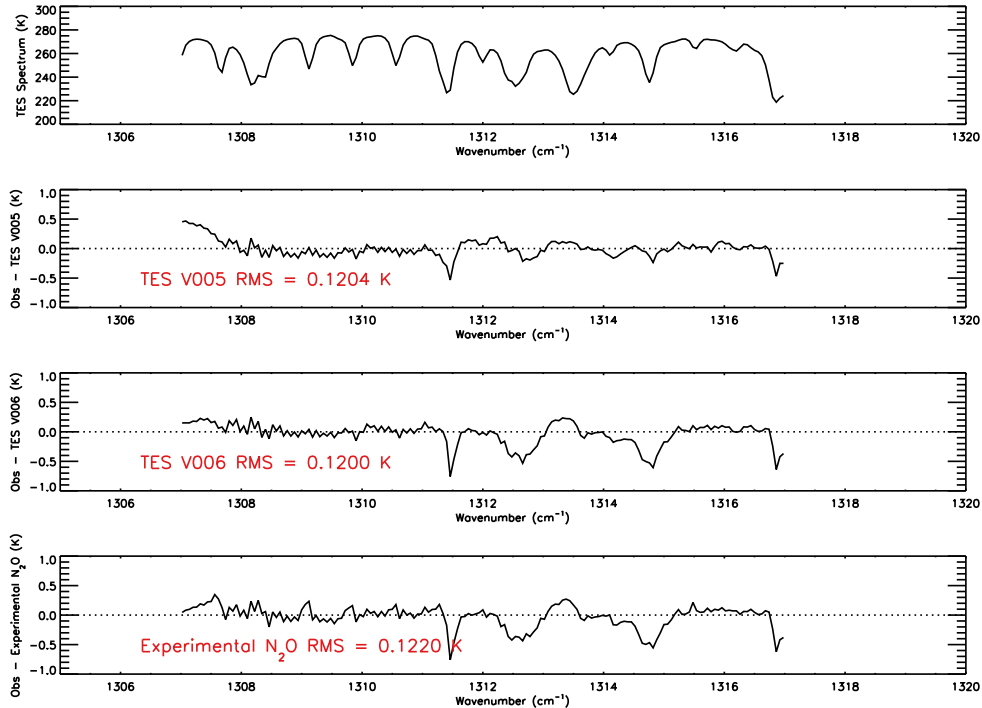


Figure R1. Mean spectral residuals as in Figure 2 of the paper, but zoomed in to the region near 1310 cm<sup>-1</sup>.

**Also, N<sub>2</sub>O<sub>5</sub> has an absorption feature near 1250 cm<sup>-1</sup>. Measuring in emission, TES can collect measurements at night when N<sub>2</sub>O<sub>5</sub> levels could be quite high. It doesn't contribute significantly in the TES measurements?**

Currently, the TES forward model does not account for the relatively weak and smooth emission feature of N<sub>2</sub>O<sub>5</sub> near 1250 cm<sup>-1</sup>, as the optical depths of H<sub>2</sub>O, CH<sub>4</sub>, N<sub>2</sub>O, and CO<sub>2</sub> are generally greater in this region. As noted above for HNO<sub>3</sub>, broad spectral features like this N<sub>2</sub>O<sub>5</sub> emission can be mitigated by the retrieval of effective cloud optical depth. We are aware of efforts to retrieve stratospheric concentrations of N<sub>2</sub>O<sub>5</sub> using infrared limb sounders like MIPAS (e.g., Mengistu Tsidu et al., 2004) and ACE-FTS (e.g., Wolff et al., 2008), but we are not aware of any studies demonstrating that this species can be retrieved in the nadir view. That said, we are also not aware of any studies that rule out such retrievals, and it is a useful suggestion for further research.

Mengistu Tsidu, G., von Clarmann, T., Stiller, G. P., Höpfner, M., Fischer, H., Glatthor, N., Grabowski, U., Kellmann, S., Kiefer, M., Linden, A., Milz, M., Steck, T., Wang, D. Y.,

and Funke, B.: Stratospheric N<sub>2</sub>O<sub>5</sub> in the austral spring 2002 as retrieved from limb emission spectra recorded by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), J. Geophys. Res., 109, D18301, doi:[10.1029/2004JD004856](https://doi.org/10.1029/2004JD004856), 2004.

Wolff, M. A., Kerzenmacher, T., Strong, K., Walker, K. A., Toohey, M., Dupuy, E., Bernath, P. F., Boone, C. D., Brohede, S., Catoire, V., von Clarmann, T., Coffey, M., Daffer, W. H., De Mazière, M., Duchatelet, P., Glatthor, N., Griffith, D. W. T., Hannigan, J., Hase, F., Höpfner, M., Huret, N., Jones, N., Jucks, K., Kagawa, A., Kasai, Y., Kramer, I., Küllmann, H., Kuttippurath, J., Mahieu, E., Manney, G., McElroy, C. T., McLinden, C., Mébarki, Y., Mikuteit, S., Murtagh, D., Piccolo, C., Raspollini, P., Ridolfi, M., Ruhnke, R., Santee, M., Senten, C., Smale, D., Tétard, C., Urban, J., and Wood, S.: Validation of HNO<sub>3</sub>, ClONO<sub>2</sub>, and N<sub>2</sub>O<sub>5</sub> from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS), Atmos. Chem. Phys., 8, 3529-3562, doi:10.5194/acp-8-3529-2008, 2008.

**Possible additional references:**

**Page 10060, line 23: Gives CH<sub>4</sub> concentrations in 1750 and 2011 without a reference. I understand the reasoning but don't like seeing quantitative values quoted without a reference.**

We agree and apologize for the oversight. The reference is Myhre et al., 2013, which is already included in the paper. We have modified our revised manuscript to make clear this is the source of these values (Page 2 Line 8). We also corrected a typo in the original values – the 1750 uncertainty should be 25 ppb, not 50 (see Page 2 Line 7).

**Page 10061, line 20: In recent years ::: Presumably this is referring to studies with GOSAT data, and it might be appropriate to provide an example reference**

We agree, and have added the following reference (see Page 2 Line 32 and Page 24, lines 26-28):

Herbin, H., Labonnote, L. C., and Dubuisson, P.: Multispectral information from TANSO-FTS instrument – Part 1: Application to greenhouse gases (CO<sub>2</sub> and CH<sub>4</sub>) in clear sky conditions, Atmos. Meas. Tech., 6, 3301-3311, doi:10.5194/amt-6-3301-2013, 2013.