

We would like to thank both referees for their time and their comments. In particular, both referees requested further discussion on the uncertainties and also how aerosols could affect TES retrievals which we address with the following discussion and paper changes.

With respect to a discussion of the uncertainties, our approach was to reference as much as possible the error discussions in previous papers so that we don't unnecessarily replicate what might be considered "boiler plate" error analysis. Therefore, we only included errors that were specific to this analysis; however we have added discussion on the expected errors of the tropospheric average of CH<sub>4</sub> and CO, used in this analysis (See Section 2.1). In addition, both referees wanted additional clarity with respect to the errors introduced by the stratosphere. We have attempted to delineate errors in the GEOS-Chem model stratosphere as well as the effect of errors in the tropopause height (and consequently the TES CH<sub>4</sub> stratospheric *a priori*) on the TES retrievals.

With respect to the role of aerosols on the TES methane retrievals, we found with the Verma et al. study that black-carbon aerosols from biomass burning have little effect on the TES retrievals for two reasons. Firstly, aerosols in biomass burning regions have been found (Kirchstetter et al., JGR 2004) to have a  $1/\lambda^2$  dependency so that an aerosol OD of 5 in the visible implies an optical depth of less than  $1e-4$  in the thermal IR. Other (unpublished literature) suggests that the dependency of this exponent on the wavelength has to be greater than 1.3. If we assume a more conservative drop-off of  $1/\lambda^{1.3}$  for the aerosol optical depth, this results in an optical depth of less than 0.01 at thermal wavelengths. Secondly, aerosols from fires have no high frequency (frequency variations  $< 1 \text{ cm}^{-1}$ ) spectral variations so that they will be effectively fitted by the estimate of cloud optical depth and cloud-top pressure (or the emissivity or surface temperature) that is co-retrieved with all TES estimates. The effect of these broad-spectral features on the TES estimates is therefore included in the overall error budget via the surface temperature, emissivity, and cloud component of the retrievals as discussed in Worden et al. (2004), Kulawik et al. 2007 and Eldering et al. 2008. This discussion on the role of clouds and aerosols is now included in the paper.

## **Response to Referee #1**

**General Comment 1:** "But for analysis of TES to compare with GEOS-Chem a truncated kernel was applied and this seems somewhat unsatisfactory. Is it not possible to use GEOS-Chem stratospheric methane profiles or some other model or instrument etc. For example in Figure 3 the *a priori* profile seems to fall off with height very rapidly above the tropopause (but we aren't shown below 1.7 ppmv of methane). Also there is a large distortion of the methane profile after the truncated profile is applied."

**Response to General Comment 1:** We have expanded the explanation for why we truncate the averaging kernels in Section 4.1. Hopefully the new explanation makes more sense! As discussed in the revised section, we ideally would account for the "smoothing error" due to the use of *a priori* information and the limited sensitivity of the TES stratospheric estimate by applying the full TES averaging kernel (surface to top-of-

atmosphere) to the GEOS-Chem model. Unfortunately, the GEOS-Chem model version that we are using has significant bias error in the middle and upper stratosphere that propagates downwards via the cross-terms in the averaging kernel to affect the comparison between the tropospheric component of data and model. We have expanded our discussion of this problem and suggested solution (truncation of the profiles for pressures lower than 80 hPa). Note that the error estimate from using this truncation approach is found to be small as discussed in the first submission of this paper. We have also added language that an update exists in which the GEOS-Chem stratospheric methane is more realistic. However, transitioning to this revised model takes some time and would also not significantly change our conclusions as the approach we use for the older model results in a small error.

Also, it is not clear what the referee means by “distortion” in the main profile. Perhaps that the model estimate after applying the TES instrument operator (averaging kernel and *a priori*) does not look like the original model field? As can be seen in Equation 1 this is a normal affect because the *a priori* and model field are different and the model estimate with the TES instrument operator applied essentially results in a linear combination of the two. If we do not apply the instrument operator to the model field then significant error between the retrieval and model is incurred because the prior and model can be significantly different. Applying the instrument operator removes these differences (e.g., Rodgers and Connor 2003).

### **General Comment 2**

“Another interesting aspect I felt uncomfortable was the issue of aerosols over the Indonesian fires...”

### **Response to General Comment 2:**

This concern is addressed in opening comments

### **General Comment 3:**

It would have been interesting to examine the methane/CO ratio for the Indonesian far outside of the region assessed with a view to assessing how long the fire signature is retained and can be measured downstream from the fire.

### **Response to General Comment 3:**

This is definitely interesting; however, oxidation of both CO and CH<sub>4</sub> as well as mixing during transport, can make the interpretation challenging. We are beginning to assimilate the TES data into the GEOS-Chem model and one of the scientific questions we wish to address is how the plume signature depends on these processes.

### **Specific Comments from Referee #1**

1) P26208 L9: I would use the word “compare” in lieu of “evaluate”. I don’t think that a model that is not driven by extensive meteorological and chemical data assimilation should be used to evaluate measurements – perhaps present sanity checks. (And even with DA in the troposphere there are issues with the PBL and convection of species).

1) Response: Done

2) P26208 L18 spelling “slope” instead of slop.

2) Response: Done

3) P26209 L14. Niggling, but I would suggest that for the Shindell et al ref an “e.g. Shindell et al” be used since this was not the first paper to discuss the idea of attacking methane and BC emissions as ones with a rapid response time, eg. Hansen et al.

3) Response: Done

4) (typo comments) P26210 L15. “Plumes . . . . .WERE (was) also observed. . . .”

P26210 L26 “methane FROM the Aura TES satellite” – word left out

P26211 L2 “TES estimates ARE mostly insensitive to . . .” word omitted.

P26213 L5 “Worden et al SHOW (no s)” plural.

P26214 L4 OH density should be  $10.8 \times 10^5$  molec  $\text{cm}^{-3}$  (105 left out)

P26216 L20 what is the rationale for doing a vertical mixing ratio average as opposed to a density average?

P26216 L26-27. Awkward wording. Suggest “becomes larger at latitudes south of  $60^\circ\text{S}$ .”

P26218 L7. Awkward wording. “Unlike (with) the methane estimates, the (this) bias in CO . . .”

P26220 L13 Award phrase. . .suggest “This correlation due to ..”

6) Response to typos: Fixed as suggested

7) Sub-heading Indonesia fire plumes, South American fire plumes etc. I think that using the word plume is misleading in the sub-headings. In doesn’t appear in the title and there is no associated plume associated with Indonesian, South American or Southern African fires. I would consider a plume as a feature well extended from the source as opposed to hovering over the source (see above)

7) Response: Changed plume to region. Section 5 title changed to  $\text{CH}_4$  and CO distributions over tropical fire plumes. I think the plural is appropriate as the observed smoke (via CO) is actually the smoke from a collection of fires and not from any one single fire.

## **Response to Referee #2**

**General Comment 1:** “It seems to me that the title of the paper is misleading since one would expect that most of the results would address the question of CH<sub>4</sub> vs. CO ...:

**Response to General Comment 1:** Prior to evaluating the distribution of atmospheric methane and CO using GEOS-Chem and TES it is useful as a sanity check to ensure that the global distribution generally agrees as disagreement would imply larger than expected errors in the data, model, or both. Global evaluation of TES data against the GEOS-Chem model is also useful for identifying errors in both data and model (e.g., poor GEOS-Chem stratosphere tropopause height) that must be addressed prior to comparison over the tropical regions discussed in the paper. While the title is not a complete representation of the paper (misleading is probably too strong a word), the overall intent of the paper is to examine tropical fires. Unfortunately, it is highly challenging to come up with a relatively short title that describes both the global comparisons and the comparisons to fires. We have however changed the title so that it emphasizes comparisons between data and model during October 2006. Suggestions are also welcome on any further title changes!

**General Comment 2:** “I agree with reviewer 1 that this paper seems to be the very first step before a more detailed study. If so, it should be clearly stated by the authors.”

**Response to General Comment 2:** We now include a motivating statement in the last paragraph of the introduction.

**General Comment 3:** “In particular, the study of the correlation during a month not affected by fires, or during other months of the fire season should be performed to confirm the results”

**Response to General Comment 3:** We have looked at fires during other months and years and in general we find that over Africa and the Amazon, larger fire emissions result in reduced slopes in the CH<sub>4</sub>/CO distributions implicating a larger role of fires on the methane distribution. However, we intend for these results to be the subject of a future paper. The focus of this paper is to characterize the comparisons between GEOS-Chem and TES data and show that we can use these data and model to examine the relative role of fires, wetlands, and transport on tropical methane distributions. We chose this time period because previous research using TES and GEOS-Chem (i.e., Logan et al. 2008 and Nassar et al. 2009) provide us with additional confidence of our results.

**General Comment 4:** “I agree with reviewer 1 that these sections should be longer. I find them very hard to follow for non-specialists (at least define what DOFS is, how are computed the various errors, etc). The definition of ‘surface-to-tropopause CH<sub>4</sub>’ should

also be stated quite explicitly. What is the justification for averaging the mixing ratios and not computing a kind of ‘tropospheric column’?

**Response to General Comment 4:** We have added language to these sections to hopefully clear up any confusion. For example, DOFS is defined but not explained. DOFS is technically the number of pieces of information. A value of 1 DOFS means that the estimate can fully resolve atmospheric variations within the calculated uncertainties for the air parcel of interest. In general, increased DOFS also results in increased vertical resolution. We choose a threshold value of 1.2 in order to ensure that the TES CH<sub>4</sub> estimate is sensitive to variations in almost the entire free troposphere (and upper boundary layer). As discussed in submitted version of the paper, we average VMR and not column amount because averaging the column would result in an estimate that is biased towards the boundary layer where the TES methane estimates shows the least sensitivity. In general though, what is most important is that the same operations that are applied to the TES data are applied to the model before the comparison in order to mitigate uncertainties due to use of *a priori* constraints and the TES vertical resolution and choice of mapping in the comparison.

**General Comment 5:** “The consequences of using truncated averaging kernels need to be better explained.”

**Response to General Comment 5:** Seen Response to Referee #1, General Comment #1.

**General Comment 6:** First, a new validation of TES CH<sub>4</sub> (version 5 with truncated kernels) with HIPPO measurements should be made to obtain the bias corresponding to the retrievals actually used here.

**Response to General Comment 6:** We are now assimilating the TES data into the latest version of the GEOS-Chem model using the approach described in this paper. The motivation for this assimilation is to re-compare the TES methane with HIPPO and also to mitigate errors from poor matchups between the TES and HIPPO observations. The results from this analysis will be in a future paper but is still too preliminary to be discussed here. However, for the fire analysis, because we subtract a global bias from the TES CH<sub>4</sub> estimate and from the GEOS-Chem CO estimate we do not expect these biases to affect our results on the correlation and slope between CH<sub>4</sub> and CO. A statement along these lines is now added in Section 4.2

**General Comment 7:** “Since the authors are mostly interested in the tropical region, they should also use measurements from CARIBIC flights: a dozen of them, measuring both CO and CH<sub>4</sub> (see for instance Schuck et al. ACP 2009 and AMT 2010) are located over Africa.”

**Response to General Comment 7:** Dr. Schuck contacted us soon after submission to let us know of their recent results using CARIBIC data. While we cannot directly compare to the CARIBIC data due to extremely different sampling both the CARIBIC and TES data show that much of the enhanced methane in plumes is likely due to wetlands or rice farming and not burning because the CH<sub>4</sub>/CO ratio in these plumes are quite large. This

conclusion is similar to our using satellite data for the plumes observed over S. America and Southern Africa and we now cite their papers.

**General Comment 8:** Then, the role played by the stratosphere is quite vague to me and I agree with reviewer 1 that it should be analyzed in more details. In particular, one subsection should be devoted to this, while the results are currently distributed in several subsections.

**Response to General Comment 8:** I think this statement is a result of our poor description of the problem in the paper. Hopefully this next iteration better clarifies the problem. Basically, We found two potential issues related to the stratosphere. The first issue is due to errors in the GEOS-Chem stratosphere and is discussed in Section 4.1. We address this error by truncating the TES CH<sub>4</sub> profile estimate and the GEOS-Chem model estimate in the lower stratosphere. A second, suspected error is potentially related to the tropopause height or to poor specification of stratospheric CH<sub>4</sub> in the TES *a priori* (Note that poor specification of stratospheric CH<sub>4</sub> results from errors in the tropopause height!) We found this error is potentially important at high latitudes but not the tropics but we include a description of this error in this paper for completeness. For these reasons we have still discussed these two errors in two separate sections but attempt to clarify the difference.

**General Comment 9:** Something that is generally missing in the paper is the study of uncertainties. What are the typical uncertainties associated to TES retrievals for both CO and CH<sub>4</sub>? How do they vary spatially and temporally, and with the thermal contrast (difference between surface and upper-air temperatures)? Thermal contrast has been shown to play a key role in both the uncertainties and DOFs that characterize the retrievals, and fire situations are particularly concerned. Also, how does it affect the averaging of TES retrievals in a GEOS-Chem grid box (for instance, are all the averaged TES retrievals in one box sensitive to the same part of the atmosphere?).

**Response to General Comment 9:** We have added explanation of the random uncertainties in Section 4.1. The effect of changing sensitivities (e.g. changing DOFS) is taken into account by applying the TES instrument operator to the geos-chem model and comparing the results before and after the operator is applied. This referee comment was highly valuable because we did not compare our results to the GEOS-chem model before the operator is applied and text is now added describing the comparison. Basically, the GEOS-Chem model has a slope of 0.11 prior to application of the instrument operator, consistent with what we expect from the CH<sub>4</sub> and CO emissions budget for Indonesia, and 0.153 afterwards. The effect of applying the operator is to include more middle and upper tropospheric air, relative to near surface air, into the comparison. However, to show that surface emissions are still the dominant feature in the comparison we now include another figure for the Indonesian fire comparison that shows the CH<sub>4</sub>/CO comparison if methane emissions from the fire are turned off. Another conclusion from this comparison is that we would expect that the emissions ratio to be smaller than 0.11 (at least 0.09) based on the difference between the GEOS-Chem and TES comparisons (Note that errors are included with the main text).

**General Comment 10:** Concerning Fig. 4, while the authors mostly study the bias, there is no discussion of the very large spread of the retrievals which does seem to vary with latitude as opposed to the bias. What is due to natural variation of methane and what is due to the instrument noise and retrieval uncertainty?

**Response to General Comment 10:** The stated errors are between approximately 8-14 ppb (as discussed in the text), therefore some of the difference is due to random uncertainties and some is due to true variability. We have added a sentence with this statement when discussing Figure 4.

**General Comment 11:** Section 5 finally deals with the study of the correlation between CH<sub>4</sub> and CO for air parcels affected by fires. In particular, some simulations where CH<sub>4</sub> from fires has been turned off are used to confirm that the slope obtained in the full comparison is characteristic of fire plumes. Here, I am wondering why the authors did not use simulations where only CH<sub>4</sub> and CO from fires are active: this would give the value expected for fire situations, to which the retrieved slope could be compared. It would seem a more direct way than turning-off CH<sub>4</sub> and finding a reduced slope.

**Response to General Comment 11:** We can actually compare directly to the emissions budget (Tables 1 and 2) for the expected ratios. The objective for turning off the emissions for methane from fires was to understand the role of transport on the comparison (both mixing and from outside the region of interest). We have added additional language pointing to Tables 1 and 2 in these sections.

**General Comment 12:** Finally, the authors state that these results ‘provide confidence in the total methane emissions from Indonesia’. I find this comment overly optimistic since it is based on one single month. Moreover, is this confidence at the level required to actually improve our understanding and estimates of the emissions themselves?

**Response to General Comment 12:** We have removed this sentence as in fact the agreement between data and model is outside 2-sigma as shown in Figure 9 and the word “confidence” implies that the comparison is within some range of uncertainties (e.g., 1-sigma, 2-sigma). The original reason for this statement was that the data and model agreed even to this level as we had expected going into this study that the differences would be much larger as it was not clear that the emissions ratios used in the model would not be sufficient for modeling the effect of these fires on the distribution of atmospheric methane.

### **Specific Comments from Reviewer #2**

1) p 26210, l. 12-14: the maps in Fig. 1 should be described to explain the general features of CO. Otherwise, it could be removed.

1) Response: We have added description explaining that the tropical enhancements in CO are related to fires.

2) p 26211, l. 3: are TES estimates insensitive to any kind of aerosols? In the thermal

infrared, it is true that fine particles like smoke don't have a strong impact on the observations. However, dust aerosols have been shown to strongly impact the observations and are even retrieved by observations from AIRS and IASI instruments.

2) Response: See general comments response. However, as noted the IR is affected by dust because of their larger size. This is not an issue for this analysis.

3) p 26212, l. 5-9: ' . . . vertical distribution of the sensitivities. . . ': is it true whatever the thermal contrast is?

3) Yes, as discussed in Worden et al. (2012).

4) p 26213, l. 13-14: the last sentence should be given earlier (Fig. 2) since it explains what TES retrievals actually are.

4) Fixed

5) p 26215, l. 22: could the authors give the averaged values of noise and temperature errors?

5) Response: We added discussion in section 2.1

6) p 26215, l. 23-26: why is the sensitivity different between CO and CH<sub>4</sub>? It should at least be illustrated.

6) Response: We have added a paragraph on CO in Section 2.1 on the vertical resolution and error characteristics.

7) p 26216, l. 18-19: could the authors give some details on how errors in the stratosphere and the tropopause height can affect the comparisons?

7) Response: See response in general comments.

8) p26216, l. 24 and 27: is it 50 or 60° S?

8) Response: We make the cutoff at 50 S but the bias is approximately the same South of 50 S but the scatter increases.

9) p 26217, l. 6-15: here the authors discuss the influence of stratosphere on CH<sub>4</sub> TES retrievals. This paragraph should be aggregated with previous sections (4.1) where it is also discussed. Are CO retrievals also affected?

9) Response: We want to keep the sections separate as they are different effects as discussed in general comments. CO retrievals are not affected as the sensitivity of the retrievals drop off in the upper troposphere, as stated in the text.

10) p 26217, l. 20: could the authors explain how the averaging is performed?

10) Response: Additional clarification provided



11) p 26217, l. 25: could the authors give some information on the spatial resolution of TES retrievals and the co-location criteria (averaged time/space distance between TES and model)?

11) We have added information about the time-step of the model in Section 3. The spatial resolution (2x2.5) and the time resolution should be sufficient information.

12) p 26218, l. 22-25: the authors now consider only some part of the retrievals. How does it affect the previous comparisons? I strongly suggest them to homogenize the retrievals used in the various sections.

12) Response: We can be more conservative in our selection of scenes in the tropics because the sensitivity (or vertical resolution) is better in the tropics due to higher temperatures. We have added a statement along these lines in the text.