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Dear Editor:

We would like to thank the referees for their time and invaluable comments.

Please see the supplement for our reply to the reviewers. Note that the referees' comments are in black and our responses are in blue.

Please contact us if there are further questions.

Best regards,
Le Kuai, PhD
Lead author
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Anonymous Referee #2

Page 6979, line 18-20: Instead of: 'It is also the OCS column Jacobians. The contour plot of OCS Jacobians (Fig. 1c) suggests that the radiances are more sensitive to OCS between 900 to 200 hPa.' May be better: 'This figure gives also the OCS column Jacobians. The contour plot of OCS Jacobians (Fig. 1c) suggests that the radiances are most sensitive to OCS between 900 to 200 hPa.'

Corrected.

page 6979, line 20: This sentence is difficult to understand. 'For comparison to the noise level, the Noise Equivalent Spectral Radiance (NESR) in this region is 1°—10−8Wcm−2 sr−1 cm−1 indicating that the OCS signal to noise ratio in this region is approximately one for each of the strong absorption lines.'

We revised the statement as below:

'Fig. 1b shows that the signal at the spectral region with strong OCS absorption is about the same or even larger than the Noise Equivalent Spectral Radiance (NESR), 1E-8 W cm^{-2} sr^{-1} cm^{-1}. Consequently, the OCS signal is detectable from the TES measured radiance with the current noise level.'

We also updated figure 1b by adding the NESR over plotted on the vertically integral Jacobian.

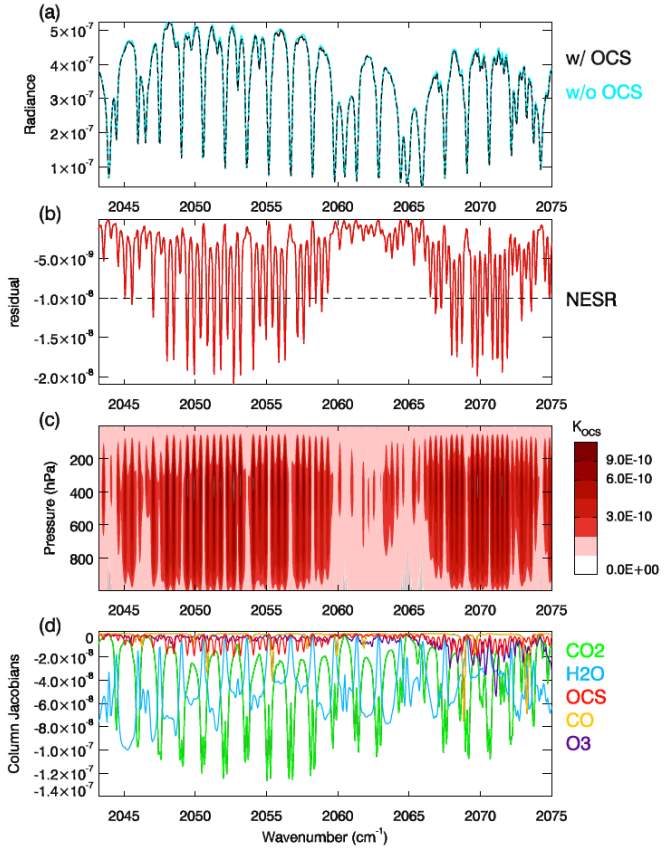

Fig. 1. (a) Model calculated radiances without OCS profile (blue line) and with OCS profile (black dash line); (b) residuals between the two models calculated radiances in (a); it is the same as OCS vertically integral Jacobians, the red line in (d); the dash line represents the Noise Equivalent Spectral Radiance (NESR), 1E-8 W cm^{-2} sr^{-1} cm^{-1}; (c) contour plot for OCS Jacobians; (d) vertically integral Jacobians for CO_2 (green), H2O (blue), OCS (red), CO (orange) and O_3 (purple).

Page 6980, line 1: The authors write: 'For these reasons, CO2 and H2O are simultaneously estimated with OCS but are constrained using estimates from a previous retrieval using different spectral bands measured by TES.' This I do not understand, are CO2 and H2O retrieved or not?

Both CO_2 and H_2O are individually retrieved step by step from their own absorption spectral regions, e.g. CO_2 from 15-micron band (Kulawik, et al., 2010). Then for the OCS retrieval, both CO_2 and H_2O are jointly retrieved (along with OCS) but with tight

constraints to their *a priori*. The a priori for CO_2 and H_2O are their previously retrieved estimates. The constraint matrices for CO_2 and H_2O are based on the covariance from their previous estimates.

Kulawik, S. S., Jones, D. B. A., Nassar, R., Irion, F. W., Worden, J. R., Bowman, K. W., Machida, T., Matsueda, H., Sawa, Y., Biraud, S. C., Fischer, M. L., and Jacobson, A. R.: Characterization of Tropospheric Emission Spectrometer (TES) CO2 for carbon cycle science, Atmos. Chem. Phys., 10, 5601-5623, doi:10.5194/acp-10-5601-2010, 2010.

This sentence is revised to:

'For these reasons, CO_2 and H_2O are simultaneously retrieved with OCS but are tightly constrained. Their a priori are estimated from previous retrieval steps, using their absorption bands that are also measured by TES (Kulawik et al., 2010; Worden et al., 2004). The constraint matrices for CO_2 and H_2O are based on the covariance from their previous estimates.'

Page 6980, line 14: The authors write: '. . . the current study is limited to over the Pacific.' This is a strong limitation, and should be named in the abstract and even title.

The title has been revised as 'Characterization of Aura tropospheric emissions spectrometer carbonyl sulfide retrievals **over ocean'**.

The first sentence in abstract has been revised to:

'We present a description of the Tropospheric Emission Spectrometer (TES) carbonyl sulfide (OCS) retrieval algorithm for **oceanic observations**, along with evaluation of the biases and uncertainties against aircraft profiles from the HIPPO campaign and data from the NOAA Mauna Loa site.'

Figure 4: It does not make sense to plot only the residuals in a Figure. The measured spectrum together with the corresponding residuals on the top must be in one Figure to make a comparison of both by eye.

We added the measured spectrum on the top of residuals, removed case 3, and rewrote the caption and discussion in the paper.

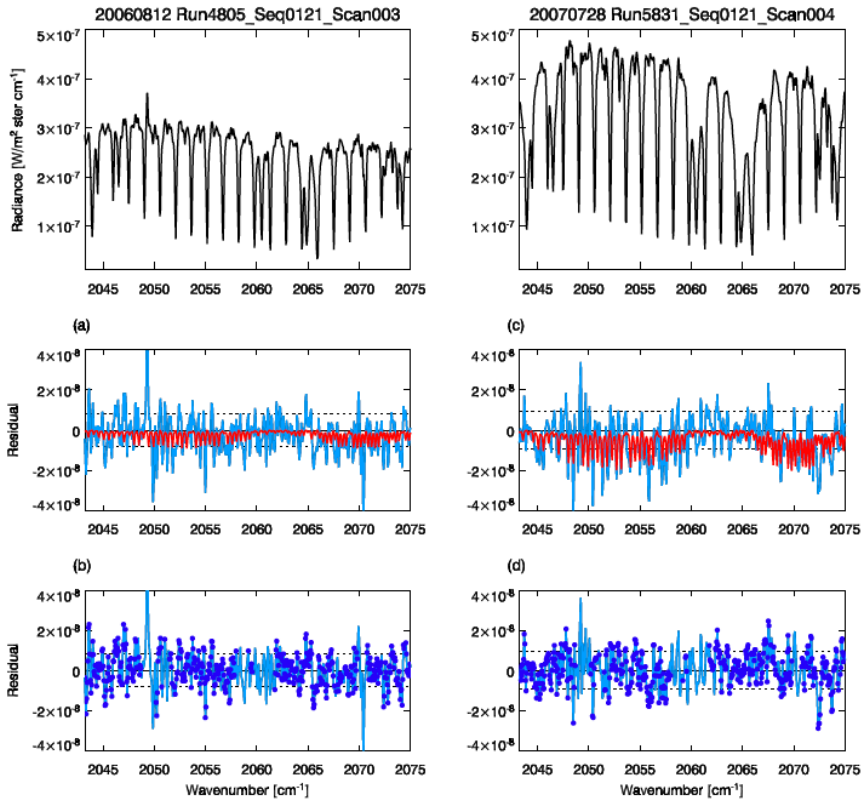

Fig. 4. Carbonyl sulfide spectral signal in the TES observations. The top two panels are the TES measured spectra for OCS retrievals. In **(a)** and **(c),** light blue lines without dots (*d*1) are the differences between TES measured spectra and forward-model run driven by retrieved CO_2 and H_2O but no OCS; red lines (*d*2) are differences between two forward-model runs with and without OCS or can be considered as OCS vertically integral Jacobians. In **(b)** and **(d)** shown in light blue lines with dots (*d*3) are the residuals after OCS retrieval. Dots indicate the frequencies of the channels used for retrieval. Solid black lines are the zero lines and dashed lines represent the noise level (NESR).

See reply for next comment for the revised discussion in the manuscript.

Page 6981, lines 10-20: I don't understand what the authors want to say in that paragraph.

This paragraph and Figure 4 are trying to show that, without OCS retrieval, the residuals between the TES measurements and model calculated spectrum is biased (see light blue lines in Figure 4 a and c). Therefore, without OCS retrieval, the residual is not only just noise but also absorption of radiation from the OCS band. However, the residuals after the OCS retrieval are much more random about zero.
We rewrote these discussions to:

'Figure 4 shows comparisons of TES radiances near Mauna Loa with modeled radiances that depend on the set of geophysical parameters affecting the observed radiance. We choose two examples of TES observations. "$d1 = \boldsymbol{y}_{\mathrm{o}} - \boldsymbol{y}_{\mathrm{m}}(\mathrm{H_2O}, \mathrm{CO_2})$" (light blue in Fig. 4a, c) is the difference between the measured TES spectra and the forward model run driven by the retrieved variables such as $\mathrm{H_2O}, \mathrm{CO_2}$ but no OCS. "$d2 = \boldsymbol{y}_{\mathrm{m}}(\mathrm{H_2O}, \mathrm{CO_2}, \mathrm{OCS}) - \boldsymbol{y}_{\mathrm{m}}(\mathrm{H_2O}, \mathrm{CO_2})$" (red in Fig. 4a, c) is the difference between the two forward model runs with and without OCS or considered as the vertically integral Jacobians of OCS. The spikes in $d1$ are related to the OCS absorption in $d2$, which suggests that without the simulation of the absorption by OCS, the residuals ($d1$) are not only the measurement noise but also the absorption by OCS ($d2$). Then, we show the residuals after the OCS retrieval ($d3 = \boldsymbol{y}_{\mathrm{o}} - \boldsymbol{y}_{\mathrm{m}}(\mathrm{H_2O}, \mathrm{CO_2}, \mathrm{OCS})$, the light blue lines with dots in Fig. 4b, d). The spikes related to OCS absorption are no longer shown in $d3$ and the residuals randomly vary about zero. Dots on $d3$ indicate the frequency of the channels selected for the retrievals.'

Page 6982, line 3: The authors write: 'Due to the low sensitivity of the TES observed radiances to OCS relative to H2O and CO2, the TES spectrum gives limited information about the OCS profile.' The retrieval of a concentration profile depends mostly on the quality of the spectral line under investigation. The relevance to H2O and CO2 is not of importance here. What do the authors want to say with this sentence?

We changed the statement to:

'Due to the low sensitivity of the TES observed radiances to OCS, the TES spectrum gives limited information about the OCS profile. Therefore, we vertically average the TES retrieval.'

Page 6982, line 9: The authors write: 'The black line is one tenth of the column averaging kernel.' I don't understand this sentence, what do the authors want to say with this sentence?

This plot intended to show that the OCS retrieval sensitivity peaks in the mid-troposphere. However, the OCS averaging kernels already demonstrate that the OCS retrieval sensitivity peaks in the mid-troposphere. We removed the black line (total averaging kernel) and the dotted line in Fig. 5 to prevent confusing readers.

Line 22: I would estimate from Figure 6 that the total error is 100 ppt (20% x 500 ppt), not 50-80 ppt.

The approximately 20% (black line in Fig. 6) for each level of the OCS profile in the free troposphere is the uncertainty of the OCS *a priori*. The total error after the retrieval (dash line in Fig. 6) is reduced to about 15% after vertically averaging in the free troposphere. The total errors of these vertically averaged OCS retrievals

ranges from 50 – 80 ppt.

We have changed the description of the errors to:

'The total error for an individual retrieval, however, is still quite large and ranges from 50 to 80 ppt, almost of the same order of magnitude for the OCS seasonal variations in the Northern Hemisphere.'

Page 6983, line 7: For me, a comparison of monthly means for many years makes no sense. In this way the result will represent the a-priori, and the procedure cannot be named comparison or validation.

The reasons we can perform a multiple-year average include (1) the inter-annual variability (no more than 10 ppt or even less) is much smaller as compared to the seasonal and latitudinal variability (more than 15 ppt or even larger), 2) the a priori is set to be a common profile for all retrievals.

We mentioned these reasons in page 6980 line 16 that

'The a priori profile of OCS is set to a constant value of 500 ppt in the free troposphere and decreases with altitude above the tropopause (Fig. 2). No obvious long-term trend is observed in atmospheric OCS, so at this stage we simply use a common OCS a priori profile to ensure that the retrieved spatiotemporal variations are not coming from the a priori.'

page 6983, line 25: The authors write: 'We attribute this result from the uncertainties in the spectroscopic line strengths.' New OCS spectral data are available for a few years, I do not agree that the 13% difference is due to uncertainties in the spectra data. Furthermore, it is not clear to me which spectral data have been used for the retrieval.

We replaced 'We attribute this result from the uncertainties in the spectroscopic line strengths.' with the statement below:

'The 13% bias could be a combination of the following effects: 1) the spectroscopic uncertainties in the OCS line parameters; 2) instrument calibration uncertainty; 3) effects of ignoring the solar contribution in the forward model; 4) errors from interfering species (e.g. CO_2, H_2O, CO and O_3). The current TES retrieval algorithm uses the forward model based on HITRAN 2008 database. The uncertainty for OCS intensities in HITRAN 2008 ranges from 2% to 20%. Attempting to quantify each of these effects is beyond the scope of this paper.'

The whole retrieval is not described in the text.

The retrieval approach is described in the section 2 (Retrieval strategy). Figure 1a shows the spectral region we use for the OCS retrieval. We mentioned on page 6979

line 12 that 'Figure 1a shows the OCS absorption spectral region from 2034 to 2075 cm−1.'

We also showed the vertically integral Jacobians of the interfering gases, such as H2O, CO2 and so on in Figure 1d and determined that (page 6979 L26)

'For these reasons, CO_2 and H_2O are simultaneously retrieved with OCS but are tightly constrained. Their a priori are estimated from previous retrieval steps, using their absorption bands that are also measured by TES (Kulawik et al., 2010; Worden et al., 2004). The constraint matrices for CO_2 and H_2O are based on the covariance from their previous estimates. CO concentrations are not jointly retrieved with OCS but are from a CO retrieval using the CO band near 2100 cm^{-1} (Worden et al., 2004).'

In section 2.1 we described the retrieval method based on the optimal estimation (Rodgers, 2000). The principle equations, such as the cost function and the estimated state vector by linear retrieval, are shown in this section. The key variables, such as Jacobians, gain matrix, and averaging kernel, are defined. Again we summarized the process of the retrieval that

'The carbonyl sulfide retrievals are carried out after the retrievals of temperature, water vapor, ozone, carbon monoxide, carbon dioxide, methane, surface temperature, emissivity, cloud optical depth, and cloud pressure (Kulawik et al., 2006). We only perform retrievals for scenes with cloud optical depth less than 0.5 as clouds reduce the sensitivity of observed radiance to atmospheric OCS. Adjustments to the atmospheric CO_2, H_2O, surface temperature, cloud optical depth, and cloud pressure are applied simultaneously with the OCS retrieval.'

Furthermore, the selection of a priori vectors and the constraint matrices are shown and discussed in section 2.3.

Page 6985, line 5: The HIPPO data must be averaged over the whole column, not just between 200 and 900 hPa. The TES data are not sensitive outside 200-900 hPa, but the a-priori used contains OCS values, which will be part of the column retrieved.

We apologize for using the term "column" in our paper when discussing the "column Jacobian" and "column averaging kernel" as this may have to assuming that the averaged OCS estimates that we discuss are a column weighted average and not a vertical average. We have changed the word column to "vertically integral (Jacobians)" or "total (averaging kernel)".

The TES retrieved OCS does not have sensitivity to the boundary layer or the stratosphere. For these reasons, we vertically average the OCS retrieval between 900 and 200 hPa.

We also used a common a priori so the a priori does not contain information of

boundary layer OCS variability. Therefore, the temporal and spatial variability of the vertically averaged OCS estimates will not depend on the *a priori*.

Page 6986, line 23: I don't understand why the bias of -15 ppt is expected. 3.4 km is still far below the tropopause.

The Mauna Loa data is a point observation at 3.4 km, a lower tropospheric measurement. While we estimate the free tropospheric OCS by averaging a profile within the mid and lower troposphere from both TES and HIPPO. Therefore, the validations using HIPPO and Mauna Loa data are slightly different and the bias should be different.

We revised the statement on page 6986, line 23 to:

'There is about −15 ppt bias, larger than the bias in HIPPO-TES comparisons, as expected because MLO data is a point measurement of the lower tropospheric OCS concentration above the boundary layer at 3.4 km instead of a profile measurement as compared to HIPPO data.'

Page 6988, line 20: Why do the authors in the conclusions give an uncertainty of 50 ppt, while on page 6982 give 50-80 ppt (and I estimate 100 ppt).

In order to make the statements about the total consistent with changes in previous comments, we revised the words as below:

'The typical uncertainties for a single observation, averaged over the troposphere, ranges from 50 to 80 ppt.'