RC1: 'Comment on amt-2021-84', Anonymous Referee #1, 07 May 2021

This is an informative and well-written paper on a subject which is important to understanding remote sensing of greenhouse gases, and should certainly be published after some revision.

Thank you for your positive comments and constructive suggestions. Please see our item-by-item responses, and associated changes (highlighted in yellow) in the revised manuscript, to your comments and suggestions below.

My suggestions and questions follow: First I will discuss two important points that I believe need explanation and a bit more consideration by the authors. Pages 8 and 9 of the manuscript describe the state vector and its relation to the underlying forward model. So for example a CO_2 a priori profile is assumed for the forward model and a single scale factor for the profile is retrieved in the state vector. For aerosol, values of AOD are retrieved for coarse and fine aerosols, and a single layer height value is retrieved. It is not at all clear how those 3 scalar quantities are related to the underlying quantities of albedo, phase function, and altitude distribution of the 5 aerosol types discussed in lines 205-224. Please explain. Are the values of CO_2 and CH_4 affected by the details of this assumed relationship?

All elements (GHG profile scale factors, AODs, and ALH) in the state vector are coupled. On the other hand, the single scattering albedos (SSAs) and phase functions of the aerosol coarse and fine modes are prescribed and not retrieved. The effective SSA for the coarse mode is calculated as the mean of the SSA values (from the GOCART model) of sea salt and dust, weighted by their simulated AODs from MERRAero. The same methodology is applied to fine-mode aerosols except using black carbon, organic carbon and sulfate. The effective phase functions can be calculated in a similar manner, except that the weighting is done by the scattering AOD.

We do not consider the geometric thickness of the aerosol layer since it has much smaller impact on the observed radiance compared to the total AOD (Zeng et al., 2019). Practically, in the forward model, the aerosols are placed in two adjacent layers. The fractions of AODs in each layer are adjusted (with total AOD conserved) to change the effective ALH. Since both fine- and coarse-mode aerosols are relatively well mixed in the atmosphere, we assume that they have the same effective ALH.

Errors in the aerosol optical properties will propagate through the forward model and affect the state vector to be optimized. To investigate this problem, a synthetic experiment (Test 3 in Section 4) has been conducted to quantify the impact on the GHG retrievals of imperfect prescription of aerosol optical properties. From this experiment, we conclude that there is no clear correlation between bias in XCO2 or XCH4 retrievals and that in aerosol optical properties of either coarse- or fine-mode aerosols. This indicates that a combination of fine- and coarse-mode aerosols is able to accurately capture the scattering effects.

We added the above paragraphs in Section 3.2.2 and Section 4.

My second important point concerns uncertainties in the retrieval. Namely, the discussion on p. 10-11 (lines 260-267), and conclusions on p. 18 attribute forward model error entirely to noise and to approximations in the calculations of multiple scattering. But surely, spectroscopic error and uncertainties in aerosol albedo and phase function are far larger. Or if not, that should be stated and justified. What has been done to evaluate these well-known sources of forward model error?

As we stated in the manuscript, ε in Equation (1) is the error vector containing both the measurement noise and the forward model error. Many previous studies have shown that the measurement noise dominates; however, the forward model error, including multiple components such as errors in spectroscopy and biases in prescribed aerosol optical properties, may not be negligible. These uncertainties propagate through the retrieval algorithm to the retrieved GHGs.

The errors due to imperfect aerosol optical properties (arising from uncertainties in aerosol fraction, size distribution, and microphysical properties) are investigated through a synthetic experiment (Test 3 in Section 4) that quantifies their impact on the GHG retrievals. As in our answer to the previous comment, from our experiment, we conclude that there is no clear correlation between bias in XCO2 or XCH4 retrievals and that in aerosol optical properties of either coarse- or fine-mode aerosols.

The benefit of using the GFIT spectroscopy database is that it has been carefully evaluated based on highly accurate TCCON observations. To further investigate the errors in spectroscopy and their impact on the GHG retrievals, we apply Principal Component Analysis (PCA) analysis on the fitting residuals. This analysis method has been used by the OCO-2/3 operational algorithm to study the effect of imperfect spectroscopy. The three principal components (PCs) with the largest variance are shown in the following figure. The features in these PCs are mostly related to spectroscopic uncertainties. These PCs might be related to line width, instrument effects, and the solar spectrum. For example, PC-3 from the WCO_2 band appears to be correlated with absorption features that can be attributed to very small changes in the line width. However, this PC can only explain a few percent of the residual variance. Overall, there are no dominant PCs that can explain more than 10% of the variance in the fitting residual. This is because the fitting residual itself is very close to random and without large systematic errors. We therefore believe that spectroscopic errors should not be a major issue here.

A summary of the above paragraphs has been added to Section 6.3 in the revised manuscript.

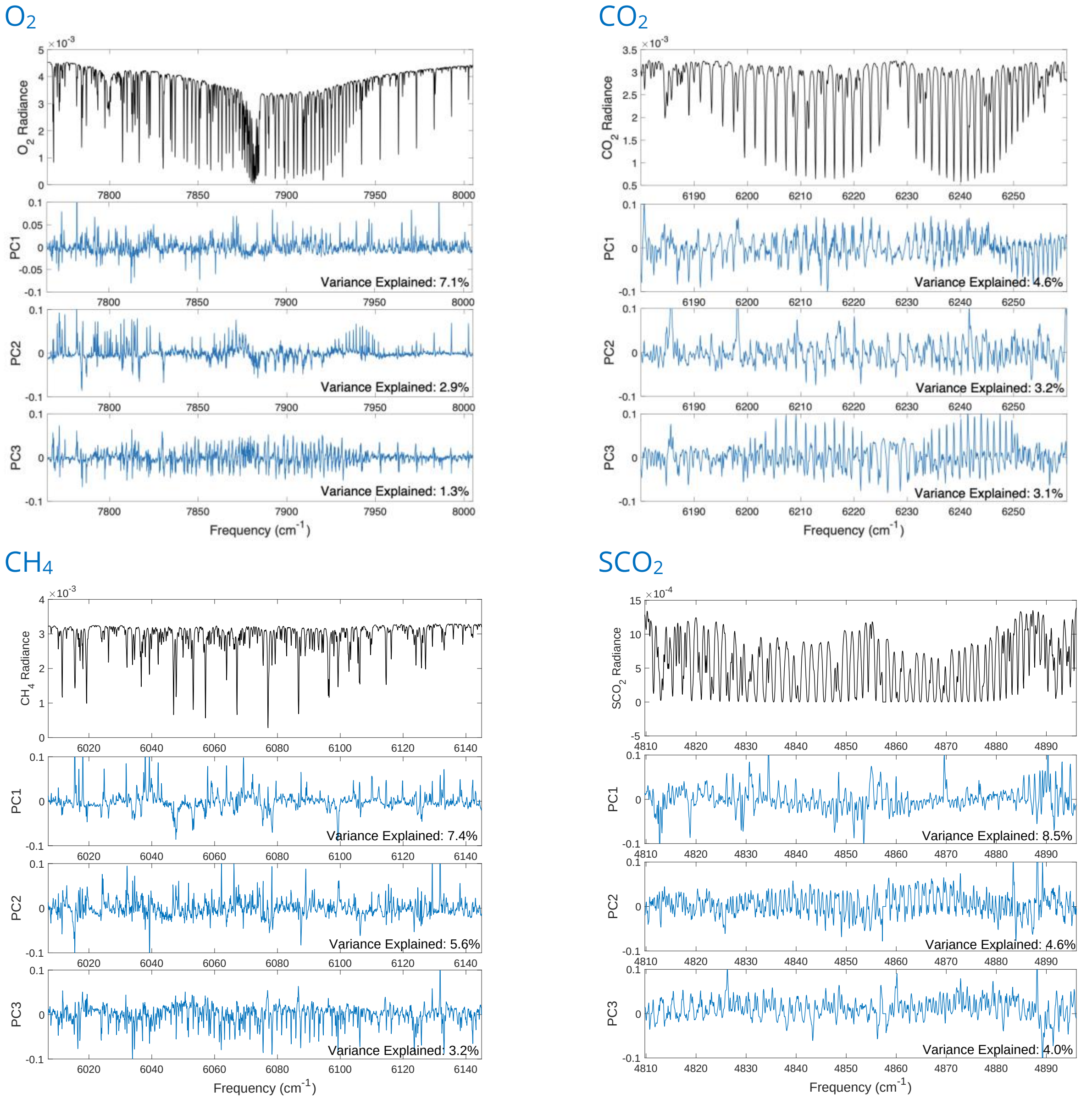

Figure A6. Mean radiance spectrum, and the three leading principal components (PCs), ranked by the variance explained by these PCs, obtained by applying Principal Component Analysis (PCA) on the fitting residuals, for the (a) O2, (b) WCO2, (c) CH4 and (d) SCO2 bands. The variance explained by each PC is also indicated.

A related point concerns the measurement error covariance matrix. The error sources I refer to are strongly correlated between channels. It is nevertheless usual in my my experience to assume a diagonal measurement covariance matrix, for practical reasons. But that assumption should be explicitly recognised as such, and the effect of those errors evaluated post retrieval.

We agree with the reviewer that correlations between channels exist and that a diagonal measurement error covariance matrix is assumed for the sake of simplicity. The correlation between channels may come from several factors, such as phase correction in converting from interferometry to radiance and detector nonlinearity. However, these impacts should be much smaller compared with measurement noise. A comprehensive evaluation of this correlation is beyond the scope of this study. However, the reduced χ^2, which is the χ^2 from equation (2) divided by the total number of measurements and state vector elements, infers the goodness of fit and can be used to evaluate the error covariance matrix. Theoretically, if the error covariance matrix is properly implemented in the retrieval algorithm, the reduced χ^2 should be close to 1 after convergence, which means that the fitting residuals are consistent with the detector noise estimates. The histogram of reduced χ^2 from all converged retrievals (see below figure) indicates that most of the retrievals have a χ^2 close to 1, with 83% having χ^2 less than 1.5. This indicates that the error covariance matrix used in the retrieval algorithm, which assumes that measurement noise is uncorrelated between spectral channels, is realistic. We also admit that inaccuracies in the spectroscopic input data and improperly modeled instrument effects may contribute to the small deviation of χ^2 from unity. Related statements have been added to Section 6.3 in the revised manuscript.

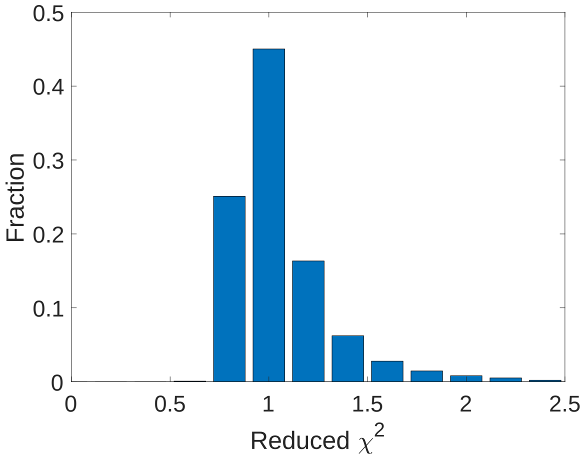

Figure A7. Histogram of reduced χ^2 from all converged retrievals in this study.

Aside from the two major points above, I have a few minor ones:

p. 2-3, lines 62-68: There is a very recent paper on GFIT2 which the authors will know about (it shares 1 co-author with this paper):

Roche, S., Strong, K., Wunch, D., Mendonca, J., Sweeney, C., Baier, B., Biraud, S. C., Laughner, J. L., Toon, G. C., and Connor, B. J.: Retrieval of atmospheric CO2 vertical profiles from ground-based near-infrared spectra, Atmos. Meas. Tech., 14, 3087–3118, https://doi.org/10.5194/amt-14-3087-2021, 2021.

This is very relevant. It has been added to the references.

p. 14, line 360, and Fig A4: The three reflecting points most used are also the closest to the instrument. I presume that explains why Fig A4 shows the AODs for other sites have mostly low values, in comparison to the AOD values for the 3 dominant sites. A sentence or two would be interesting to confirm and interpret that.

We add the following sentences in Section 5.3.

In comparison to the three sites close to the CLARS location (Santa Anita, Santa Fe, and West Pasadena), for sites that are further away, valid retrievals that pass the filters have lower AOD values. This is because of their longer slant paths in the PBL, leading to a larger scattering effect even under the same vertical aerosol loading.

p. 15, line 397: CLARS-FP?

Corrected. It should be GFIT3.

p. 17: Section 6.2 is confusing. The discussion jumps back and forth from one assumption to another about reflectance ratio, and loses this reader.

We have substantially rephrased this paragraph. Please see Section 6.2 in the revised manuscript.