Response to Reviewer Comments

We truly appreciate the reviewer#3 for the thoughtful and helpful comments. Below are our responses (in regular font) to the reviewer's comments (in ***bolded italic*** font).

Reviewer #3:

The manuscript investigates the retrieval of GHGSat-like methane retrievals in the presence of a simulated aerosol layer. Performance of the retrieval is investigated in four different configurations: $\Delta XCH4$ only in nadir and multi-angle viewing methods, and simultaneous $\Delta XCH4$ and AOD in both viewing approaches. Bias and standard deviation in the retrieval are investigated under varying satellite viewing angles, aerosol SSA, asymmetry factor (g), and surface albedo. Some aspects of the work do feel simplistic, as described by Referee #1. However, the results are clearly described, and the interaction with aerosols is important to understand in the satellite methane community. I believe the work is suitable for publication, if the following comments are addressed:

Regarding the notation "X_{AOD}" used in the text, this choice seems unusual. The numerical plots already simply use "AOD", which I would encourage.

- X_{AOD} has been replaced as AOD in the manuscript.

L9 'polarity of ΔXCH_4' – suggest rephrasing as the sign of ΔXCH_4 biases

- Corrected.

L104 "In Fig. 1b, strong CH_4 absorptions are found around 1666 nm, affirming that the DISORT-simulated radiance is adequate for simulating the methane effect." This conclusion is of importance but seems a little weak. Can it be strengthened with more evidence, or past validation?

- GHGSat measures methane concentrations by analyzing the spectrally decomposed solar backscattered radiation within the methane absorption band (~1.65μm) (Jervis et al., 2021). If DISORT can simulate radiance identical to GHGSat and capture the methane spectra absorption features when given specific atmospheric profiles and CH_4 mixing ratio, it can effectively mimic the GHGSat measurement. The methane absorption features observed in TOA reflectance, as demonstrated in our paper in Figure 1b, align closely with results from other studies, such as Figure 3a in (Jervis et al., 2021) and Figure 2 in (Chan Miller et al., 2023). This consistency suggests the robustness and validity of our radiance simulations. In our study, we combine the LBLRTM, DISORT, and GHGSat instrument models as the

forward model to simulate GHGSat measurements. Following the reviewer's suggestion, discussions have been added at Lines 119-122.

L134 "solar and satellite zenith angles" is repeated twice. The second occurrence should be azimuth

- Corrected.

Fig 2 It would be preferable to draw the phi2 angle to the projected point (possibly lying on the x-axis), not to the dashed backward viewing line. Often, these figures are labeled with North/South. Additionally, I am now realizing that this figure does not show the same set of angles as Table 1, which seems a little confusing, though not necessarily 'wrong' since it is a schematic diagram.

- Thank you for the suggestions. Figure 2 has been redrawn completely.

I would like to understand the difference between Fig 3 and Fig 1(c). The main two differences seem to be: the 0.3 FWHM smoothing and the added simulation of the 0.1 AOD aerosol layer. Looking at regions such as 1664-1665 nm, I do not think the coarser resolution explains the quantitative differences of values near ~0.2 compared to ~0.15. Is this then explained by the aerosol layer, or also other factors?

- Thank you for the comment. In Figure 3, we added sulfate aerosols with an AOD of 0.1 in the DISORT simulation. The results in Figure 3 have been updated. The differences in simulated reflectance between Figure 3 (green line, nadir view) and Figure 1(c) are purely attributed to aerosols. As shown below in Fig.S1, we have plotted the simulated GHGSat TOA reflectance for scenarios of clean atmospheric conditions and AOD 0.1 for comparison. Sulfate aerosols introduce more atmospheric scattering and result in a slightly higher TOA reflectance. The magnitude of aerosol-induced TOA reflectance change is around 10^{-3}. Relevant discussions are added at Lines 197-200.

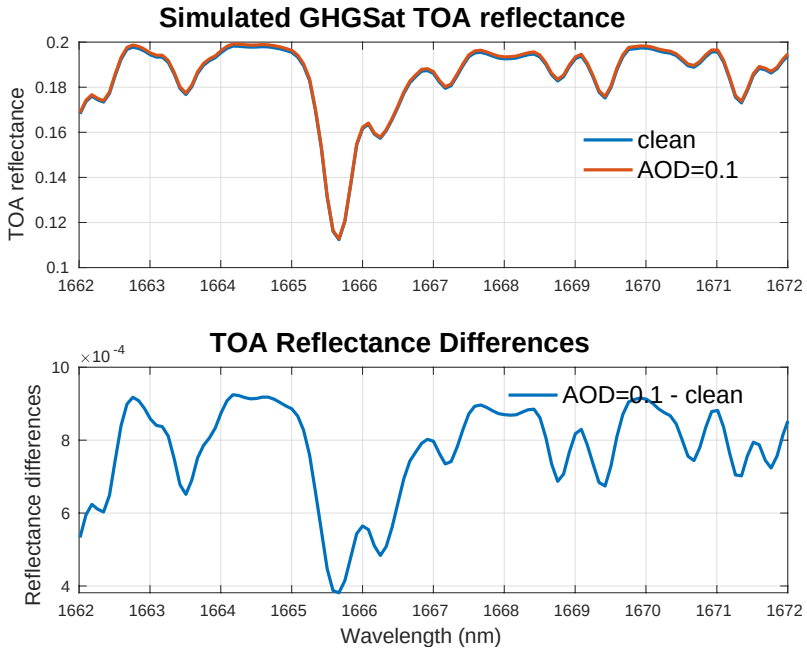

Figure S3. Upper: Simulated TOA reflectance measured by GHGSat instrument at a spectral resolution of 0.3 nm FWHM for clean condition and AOD = 0.1 condition. Bottom: Reflectance differences between AOD =0.1 condition and clean condition. The instrument observes the surface with an albedo of 0.2 at nadir viewing positions. For the AOD condition, sulfate aerosols with 0.1 AOD at SWIR are added near the surface.

In the title of Figure 3, what is SU in AOD(SU)?

- SU stands for sulfate aerosols. The title and caption of Figure 3 have been changed accordingly.

In Figure 4, can the first step (labeled DISORT), be a little descriptive, to distinguish it from DISORT in the forward model step?

- Thank you for the suggestion. Figure 4 has been updated.

L216-217 Note that Fig 6b (phase function for specific g) is not explicitly mentioned, and the link could be clearer to what is discussed (intensity of scattering energy)

- Descriptions of Fig 6b have been added at Lines 276-279.

L229 I do not necessarily understand the uncertainty simulation here. Is 0.2% the combined magnitude of the white noise and 1/f noise, or for each individually? Is uncertainty of the aerosol optical properties included somehow?

- In response to the reviewer's comment about the 0.2% magnitude of noise, we introduced white noise and 1/f errors, each with a standard deviation of 0.2%, to account for instrument measurement uncertainty. These settings are considered reasonable within the GHGSat system. Clarifications are added at Line 298.

- Regarding the reviewer's comment on the uncertainties introduced by aerosol optical properties, we have added results in Section 3.2. We assumed certain aerosol SSA, g, and height distributions in retrieval (e.g. Jacobian calculation), while for the simulated GHGSat radiance, we incorporated more complex representations for aerosol type and height distributions. The differences between retrieval with fixed (inaccurate) parameters and retrieval with real (accurate) parameters enable us to quantify the uncertainty resulting from the inaccurate representation of these parameters.

Aerosol Type Uncertainties

Fig. S2 presents the differences in mean bias and standard deviations of retrieved variables between retrievals assuming SSA = 0.95 and g = 0.7 for aerosols and retrievals assuming the correct SSA and g. These differences could suggest the uncertainty of simultaneous retrieval when assuming inaccurate aerosol types. Fig.S2a and S2d show that the uncertainty in the mean bias and STD of ΔX_{CH_4} related to aerosol types ranges from -5.8% to 2.7% and -0.2 to 0.9%, respectively, for typical aerosol optical property values. The uncertainty in the mean bias and STD of AOD falls within -40.2% to 16.1% and -9.6% to 20%, respectively. Similarly, the uncertainty in the mean bias and STD of X_{alb} ranges from -5.6% to 5.4% and -1.5% to 0.39%, respectively. These findings suggest that even with incorrect SSA and g assumptions in the retrieval, the maximum uncertainty induced in the accuracy of retrieved ΔX_{CH_4} is within 5.8%.

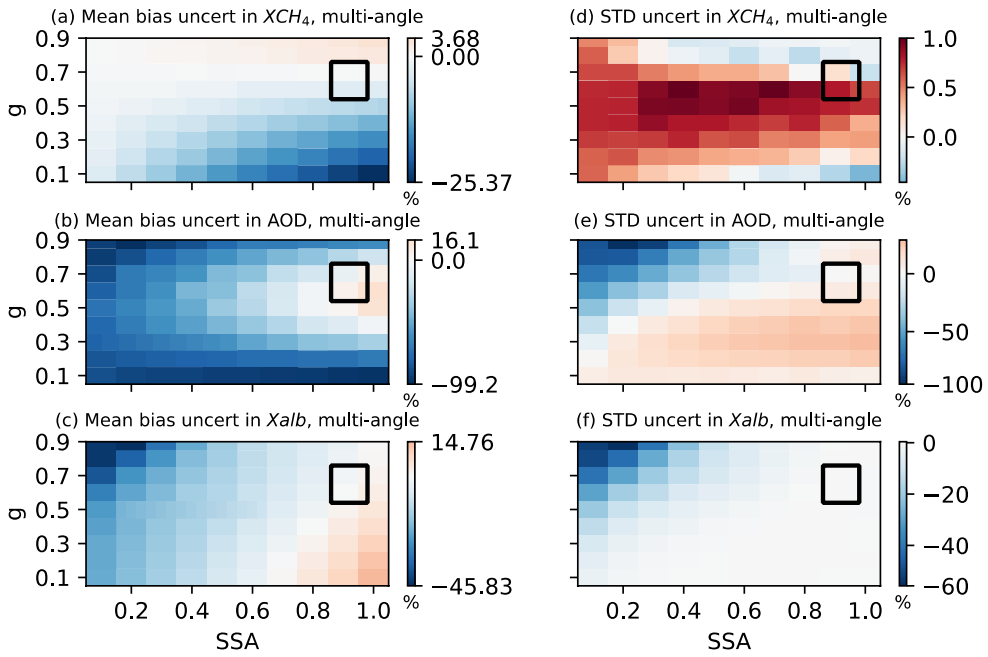

Figure S2. Uncertainties induced by aerosol type in mean bias (left column) and standard deviations (STD) (right column) of retrieved ΔX_{CH_4}, AOD, and X_{alb}, assuming aerosols with an SSA of 0.95 and a g of 0.7 in the retrieval. The simulated truth of ΔX_{CH_4}, AOD, and X_{alb} are 0.1 ppm, 0.1, and 0.2, respectively. The scattering angle ranges from 100°-140°. The black box represents the typical values for aerosol optical property ranges (SSA ∈ [0.86, 0.98] and g ∈ [0.54, 0.76]) in the observation.

Aerosol Height Distribution Uncertainties

While aerosols primarily reside near the surface at the industrial site, they could also ascend to higher altitudes under favorable atmospheric conditions. Therefore, we examined the uncertainty brought by aerosol height assumptions. We compared the differences between the retrieval when we assume aerosols are near the surface with those when aerosols are elevated to 5 km. In the latter case, AOD linearly decreases with height but we still use the near-surface Jacobian calculations in retrieval. Fig.S3 shows the uncertainties in simultaneous retrieval when assuming incomplete aerosol height.

Similar to the uncertainty results related to aerosol types, Fig.S3a and S3d show that the uncertainty induced by aerosol height in the mean bias and STD of ΔX_{CH_4} ranges from 2.3% to 6.4% and from -0.1 to 0.1%, respectively, for typical values of aerosol optical

properties. The mean bias uncertainty for AOD and X_{alb} falls within the range of 2.3% to 41.5% and -0.8 to 1.4%, respectively. The STD uncertainty for ΔX_{CH_4}, AOD, and X_{alb} is generally small, indicating minimal sensitivity of retrieval precision to the aerosol height distributions.

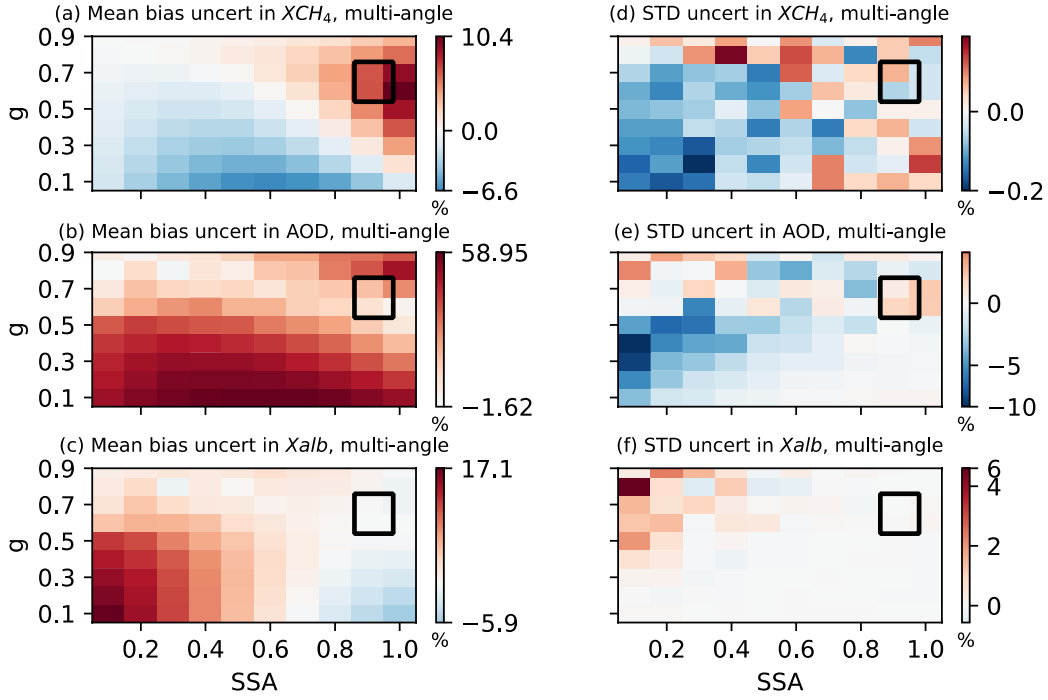

Figure S3. Uncertainties induced by aerosol height distributions in mean bias (left column) and standard deviations (STD) (right column) of retrieved ΔX_{CH_4}, AOD, and X_{alb}, assuming near-surface aerosols in the retrieval. The simulated truth of ΔX_{CH_4}, AOD, and X_{alb} are 0.1 ppm, 0.1, and 0.2, respectively. The scattering angle ranges from 100°-140°. The black box represents the typical values for aerosol optical property ranges (SSA ∈ [0.86, 0.98] and g ∈ [0.54, 0.76]) in the observation.

In summary, the uncertainty in the mean bias and STD of ΔX_{CH_4} induced by inaccurate aerosol types and height distributions is less than 6.4% and 0.9%, respectively. This uncertainty is obtained when assuming near-surface aerosols with fixed SSA (0.95) and g (0.7) and a 0.2 surface albedo in retrieval, while in simulated radiance, aerosol SSA, g, and height distribution vary across typical observation ranges.

L356 'A bunch of' – Suggest to quantitatively specific the satellite zenith angles considered

- Corrected.

While satellite zenith angles were considered, what about solar zenith angle? I believe this was fixed to 60 degrees throughout and wonder how this influences the results.

- Adjusting the solar zenith angle gives us retrieval results similar to those discussed in section 4.1. When we alter the solar and satellite zenith angles, the scattering angle changes, thereby influencing our retrieval. In section 4.1, we have examined extremely wide scattering angle ranges (40° ~ 180°), which are likely among the most extreme values seen in real observations. Figure 12 indicates that simultaneous methane and aerosol retrievals exhibit relatively small mean bias when the satellite zenith angle (scattering angle range) is small. This suggests that there is little requirement for the angle setting when applying the multi-angle viewing method to the GHGSat instrument.

Figure 8 and others, note there is a discrepancy between the figure ("Mean bias in XCH4") and caption (Mean bias of ΔXCH_4)

- Thank you for the comments. All captions are updated accordingly.

References

Chan Miller, C., Roche, S., Wilzewski, J. S., Liu, X., Chance, K., Souri, A. H., Conway, E., Luo, B., Samra, J., Hawthorne, J., Sun, K., Staebell, C., Chulakadabba, A., Sargent, M., Benmergui, J. S., Franklin, J. E., Daube, B. C., Li, Y., Laughner, J. L., Baier, B. C., Gautam, R., Omara, M., and Wofsy, S. C.: Methane retrieval from MethaneAIR using the CO_2 Proxy Approach: A demonstration for the upcoming MethaneSAT mission, Gases/Remote Sensing/Data Processing and Information Retrieval, https://doi.org/10.5194/egusphere-2023-1962, 2023.

Jervis, D., McKeever, J., Durak, B. O. A., Sloan, J. J., Gains, D., Varon, D. J., Ramier, A., Strupler, M., and Tarrant, E.: The GHGSat-D imaging spectrometer, Atmos. Meas. Tech., 14, 2127–2140, https://doi.org/10.5194/amt-14-2127-2021, 2021.