

Dear Editor,

We would like to thank you and the two reviewers for your constructive comments and suggestions to improve the clarity of our manuscript. We have made changes to address these comments and suggestions. The following are the main changes:

1. More information has been added to the abstract to increase clarity, quantify the results better and summarize the comparison between the MF and OE methods.
2. Details have been added about the treatment of aerosols and surface albedo.
3. Explanations are supplied for the behavior of the retrievals as a function of the different parameters.
4. More quantification is provided for the differences between the two retrieval techniques.
5. Effects of changing the *a priori*, *a priori* error and simulation spectral resolution are described.
6. New Tables and Figures have been added to provide more detail.

Point-by-point responses to the comments are provided below. The reviewer comments are in blue, our responses are in red (line numbers refer to those in the revised manuscript), and modifications to the original manuscript are highlighted in yellow.

Vijay Natraj

On behalf of all co-authors

This paper studies the impact of aerosols on methane retrievals from synthetic AVIRIS-NG-like measurements using two retrieval methods, the traditional match filter (MF) method, and the more modern and quantitative optimal estimation (OE) method that uses radiative transfer model and can include more physics. It shows how both retrievals are sensitive to various AOD, CH<sub>4</sub> concentration, surface albedo, SSA and g. The scope of this study is well suited for AMT. This paper is generally well organized and methodology is generally good. However, a lot of results are shown without much further explanation about the physics behind. Some of the statements may be questionable due to different definitions of retrieval biases between the two retrievals. Some key information (e.g., surface albedo treatment in OE retrieval) is not clearly described. The abstract also needs to be improved as it does not clearly summarize this study. Overall, I think that this paper can be published after addressing the specific comments below.

We thank the reviewer for the encouraging words and for the excellent suggestions. We have added/modified some sentences (lines 24–40) in the abstract to more clearly summarize the study.

Using a numerically efficient two-stream-exact-single-scattering radiative transfer model, we also simulate AVIRIS-NG measurements for different scenarios and quantify the impact of aerosol scattering in the two retrieval schemes by including aerosols in the simulations but not in the retrievals. The presence of aerosols causes an underestimation of CH<sub>4</sub> in both the MF and OE retrievals; the biases increase with increasing surface albedo and aerosol optical depth (AOD). Aerosol types with high single scattering albedo and low asymmetry parameter (such as water soluble aerosols) induce large biases in the retrieval. When scattering effects are neglected, the MF method exhibits lower fractional retrieval bias compared to the OE method at high CH<sub>4</sub> concentrations (2–5 times typical background values), and is suitable for detecting strong CH<sub>4</sub> emissions. For an AOD value of 0.3, the fractional biases of the MF retrievals are between 1.3 and 4.5%, while the corresponding values for OE retrievals are in the 2.8–5.6% range. On the other hand, the OE method is an optimal technique for diffuse sources (<1.5 times typical background values), showing up to five times smaller fractional retrieval bias (8.6%) than the MF method (42.6%) for the same AOD scenario. However, when aerosol scattering is significant, the OE method is superior since it provides a means to reduce biases by simultaneously retrieving AOD, surface albedo and CH<sub>4</sub>. The results indicate that, while the MF method is good for plume detection, the OE method should be employed to quantify CH<sub>4</sub> concentrations, especially in the presence of aerosol scattering.

The treatment of aerosols and surface albedo is now described in lines 172–179.

Aerosols are not included in the state vector for both the real and synthetic retrievals. They are, however, considered in the forward model for the synthetic simulations. Table 1 lists optical properties for four basic aerosol types (dust, water soluble, oceanic and soot). Table 2 shows the corresponding properties for three aerosol models that are defined as mixtures of the basic components from Table 1 (WCRP, 1986). We employ the Henyey-Greenstein phase function (Henyey and Greenstein, 1941), where aerosol composition is determined by two parameters: single scattering albedo (SSA) and asymmetry parameter ( $g$ ). The surface albedo is also not retrieved; for both real and synthetic retrievals, it is held fixed and assumed to be independent of wavelength.

Further description of the retrieval bias as a function of SSA and  $g$  is provided in lines 302–312.

This behavior can be explained as follows. At higher SSA values, there are more multiple scattering effects (that are ignored in the retrieval). On the other hand, larger values of  $g$  imply greater anisotropy of scattering (preference for forward scattering), leading to a reduction in multiple scattering effects. Since the retrieval bias is large for high SSA and low  $g$ , the water-soluble aerosol type (Table 1) and the maritime aerosol model (Table 2) can be expected to induce greater biases in the retrieval. In order to compare the impacts

of SSA and  $g$  in further detail, retrieval results due to a  $\pm 5\%$  change in SSA and  $g$  for the three aerosol models from Table 2 are shown in Figures 7b and 7c. Note that for the maritime aerosol model, the SSA is set to 0.999 for the +5% scenario to ensure physicality. It is clear that (1) the maritime aerosol model induces larger retrieval biases than the other aerosol types, and (2) the retrieval results are more sensitive to changes in  $g$  than those in SSA.

The effects of changing the *a priori*, *a priori* error and RT simulation spectral resolution are described in lines 325–334.

The effects of changing the *a priori*, *a priori* error and RT simulation spectral resolution on the retrieved XCH<sub>4</sub> are shown in Figure 9. For these calculations, the other parameters are set as follows: SSA = 0.95,  $g = 0.75$ , AOD = 1.0, surface albedo = 0.5 and true XCH<sub>4</sub> =  $5.8 \times 1.822$  ppm. The parameters were chosen to correspond to the scenario with the largest retrieval bias in Figure 8c (bottom right box in Figure 8c). Figure 9a shows that the retrieved XCH<sub>4</sub> changes by about 9 ppb as the *a priori* changes from half to twice the true XCH<sub>4</sub> value. Similarly, the XCH<sub>4</sub> difference is less than 4 ppb when the *a priori* error changes from 0.05 to 0.5 (Figure 9b). Compared to the bias of about 923 ppb induced by neglecting aerosol scattering for this scenario, it is clear that the impacts of the *a priori* and *a priori* error are very small. The effect of spectral resolution is larger, but XCH<sub>4</sub> still changes by only about 100 ppb when the spectral resolution is changed from 0.5 to 0.1 cm<sup>-1</sup> (Figure 9c).

We provide more quantification of the differences between the two retrieval techniques (lines 339–351).

The bias ratio for the MF method (1.3–4.5%) is up to 53.6% less than that for the OE method (2.8–5.6%) for AOD = 0.3 when the CH<sub>4</sub> concentration is high (2–5 times typical background values). On the other hand, the OE method performs better when enhancements are small and XCH<sub>4</sub> is close to the background value. For example, the bias ratio for the MF method has a high value of about 42.6% at AOD = 0.3 for a 10% enhancement (XCH<sub>4</sub> =  $1.1 \times 1.822$  ppm); the OE value for the same scenario is 8.6%. For scenarios where scattering is ignored, the two retrieval techniques seem to be complementary, with differing utilities for different enhancements. On the other hand, when RT models that account for scattering effects are employed, the MF technique is suboptimal. Further, MF retrievals rely on accurate characterization of the surface albedo, especially when the aerosol loading is large. Finally, the MF method does not retrieve concentrations, which are necessary to infer fluxes. Therefore, the OE technique is in general superior due to its ability to support simultaneous retrieval of aerosols, surface albedo and CH<sub>4</sub> concentration.

Finally, we have added two new Tables (Tables 1 and 2) and several new Figures (Figures 7b, 7c, 8d and 9).

#### Specific comments

1. Subscripts and superscripts in the text are disproportionately too small to read.

The subscripts and superscripts look fine in our version. Perhaps this is an issue that could be addressed by the journal publication team after acceptance.

2. L19-21, this sentence is not consistent with the text as real AVIRIS-NG data are mainly used to compare both MF and OE retrievals, rather than analyze the impact of aerosol scattering on CH<sub>4</sub> retrievals. That is probably why the first reviewer commented that section 3 is loosely connected with the main purpose of this paper. I suggest clearly describing the purpose of this section 3 probably in the introduction section in relation to the main topics of this study.

The reviewer is right. We modified the abstract to indicate the use of AVIRIS-NG data (lines 21–24).

In this study, imaging spectroscopic measurements from the Airborne Visible/Infrared Imaging Spectrometer–Next Generation (AVIRIS-NG) in the short-wave infrared are used to compare two retrieval techniques — the traditional Matched Filter (MF) method and the Optimal Estimation (OE) method, which is a popular approach for trace gas retrievals.

We also describe the organization of this work at the end of the introduction (lines 113–118).

This article is organized as follows. The MF and OE retrieval methods are described in Section 2. Section 3 focuses on analysis of a sample CH<sub>4</sub> plume detected by AVIRIS-NG measurements and compares retrievals using the MF and OE methods. Section 4 presents a detailed evaluation of aerosol impacts on the two retrieval methods through simulations of AVIRIS-NG spectra for different geophysical parameters. Section 5 provides a summary of the work and discusses future research.

3. In abstract, the sentence in L25-29, it is not clear about what kind of scenario for the retrieval bias. Please make it clear here that this for retrieval underestimation of CH<sub>4</sub> when aerosol is present but neglected in the retrieval. The use of 50% here and also in the text in Section 4.4 is very confusing especially you have >100% enhancement and < 50% enhancement and also the retrieval bias is actually on the order of 1.5-6%. You may use

something like “half of the retrieval bias” or provide specific retrieval bias ( $\sim 2\text{-}6\%$  for OE and  $1.5\text{-}3.5\%$  for MF) based on Figure 9. The sentence is also too long. You may rephrase it in a couple of sentences.

We thank the reviewer for the excellent suggestions. We now indicate that aerosols are present in the simulations but neglected in the retrieval (lines 24–27).

Using a numerically efficient two-stream-exact-single-scattering radiative transfer model, we also simulate AVIRIS-NG measurements for different scenarios and quantify the impact of aerosol scattering in the two retrieval schemes by including aerosols in the simulations but not in the retrievals.

We have also rephrased the sentences comparing the two retrievals (lines 30–37).

When scattering effects are neglected, the MF method exhibits lower fractional retrieval bias compared to the OE method at high  $\text{CH}_4$  concentrations (2–5 times typical background values), and is suitable for detecting strong  $\text{CH}_4$  emissions. For an aerosol optical depth (AOD) value of 0.3, the fractional biases of the MF retrievals are between 1.3 and 4.5%, while the corresponding values for OE retrievals are in the 2.8–5.6% range. On the other hand, the OE method is an optimal technique for diffuse sources ( $<1.5$  times typical background values), showing up to five times smaller fractional retrieval bias (8.6%) than the MF method (42.6%) for the same AOD scenario.

We add two sentences to summarize the implications of this work (lines 37–40).

However, when aerosol scattering is significant, the OE method is superior since it provides a means to reduce biases by simultaneously retrieving AOD, surface albedo and  $\text{CH}_4$ . The results indicate that, while the MF method is good for plume detection, the OE method should be employed to quantify  $\text{CH}_4$  concentrations, especially in the presence of aerosol scattering.

We have also provided further details in Section 4.4 (lines 339–351).

The bias ratio for the MF method (1.3–4.5%, AOD = 0.3) is up to 53.6% less than that for the OE method (2.8–5.6%, AOD = 0.3) when the  $\text{CH}_4$  concentration is high (2–5 times typical background values). On the other hand, the OE method performs better when enhancements are small and  $X\text{CH}_4$  is close to the background value. For example, the bias ratio for the MF method has a high value of about 42.6% at AOD = 0.3 for a 10% enhancement ( $X\text{CH}_4 = 1.1 \times 1.822$  ppm); the OE value for the same scenario is 8.6%. For scenarios where scattering is ignored, the two retrieval techniques seem to be

complementary, with differing utilities for different enhancements. On the other hand, when RT models that account for scattering effects are employed, the MF technique is suboptimal. Further, MF retrievals rely on accurate characterization of the surface albedo, especially when the aerosol loading is large. Finally, the MF method does not retrieve concentrations, which are necessary to infer fluxes. For these cases, the OE technique is superior due to its ability to support simultaneous retrieval of aerosols, surface albedo and CH<sub>4</sub> concentration.

4. In abstract, L29-31, it is good to summarize main results instead of just describing what are discussed.

We now summarize the main results per the reviewer's suggestion (lines 27–37).

The presence of aerosols causes an underestimation of CH<sub>4</sub> in both the MF and OE retrievals; the biases increase with increasing surface albedo and aerosol optical depth (AOD). Aerosol types with high single scattering albedo and low asymmetry parameter (such as water soluble aerosols) induce large biases in the retrieval. When scattering effects are neglected, the MF method exhibits lower fractional retrieval bias compared to the OE method at high CH<sub>4</sub> concentrations (2–5 times typical background values), and is suitable for detecting strong CH<sub>4</sub> emissions. For an AOD value of 0.3, the fractional biases of the MF retrievals are between 1.3 and 4.5%, while the corresponding values for OE retrievals are in the 2.8–5.6% range. On the other hand, the OE method is an optimal technique for diffuse sources (<1.5 times typical background values), showing up to five times smaller fractional retrieval bias (8.6%) than the MF method (42.6%) for the same AOD scenario.

5. L74, suggest changing "large" to "large number of " as it implies coarser spatial resolution contrary to "fine spatial resolution".

We have made the suggested revision (line 85).

6. L78, suggest changing to "a spectral resolution of 5 nm full width at half maximum (FWHM)"

We have made the suggested revision (line 89).

7. At the end of the introduction, it would be useful to add how this paper is organized in following sections.

We now describe the organization of the paper (lines 113–118).

This article is organized as follows. The MF and OE retrieval methods are described in Section 2. Section 3 focuses on analysis of a sample CH<sub>4</sub> plume detected by AVIRIS-NG measurements and compares retrievals using the MF and OE methods. Section 4 presents a detailed evaluation of aerosol impacts on the two retrieval methods through simulations of AVIRIS-NG spectra for different geophysical parameters. Section 5 provides a summary of the work and discusses future research.

8. Units on both sides of Equation (5) do not match. According to the text, V has a unit of liter / mol or 1E-3 m<sup>3</sup>/mol, and 1/(V\*1E3) has a unit of mol/m<sup>3</sup>. Maybe 1/(V\*1E3) should be V\*1E-3 instead. Or V has a unit of mol/liter, then it should not be called V as it is confusing. So please clarify this.

Good catch. Not only was the equation wrong, but it was also confusing. The equation now reads (line 147):

$$\kappa_{\text{traad}} [\text{m}^2 \cdot \text{mol}^{-1}] = \kappa [\text{ppm}^{-1}\text{m}^{-1}] \times V [\text{liter mol}^{-1}] \times 10^{-3} [\text{m}^3 \text{liter}^{-1}] / 10^{-6} [\text{ppm}^{-1}] \quad (5)$$

$\kappa_{\text{traad}}$  is defined in line 142 as the absorption coefficient in units of  $\text{m}^2 \cdot \text{mol}^{-1}$ .

9. L132-135, the sentence does not read well here as the purpose of using real AVIRIS-NG data has not been introduced yet. You may rephrase it to something like “To illustrate the MF retrieval and its difference from the OE method, we perform MF retrievals from AVIRIS-NG measurement made on ... as shown in Fig. 2. The samples for...” Or it might be even better to move these two sentences to Section 3 before showing MF retrieval results.

The reviewer is right. We modified the first sentence (lines 149–150).

The background mean radiance  $\mu$  used in Equation 4 is based on the AVIRIS-NG measurement shown in Figure 2; this is described in more detail in Section 3.

As recommended by the reviewer, we added a sentence at the start of Section 3 (lines 182–184) to introduce the purpose of using AVIRIS-NG data.

To illustrate the OE retrieval and its difference from the MF method, we perform retrievals for an AVIRIS-NG measurement made on 4 September 2014 (ang20140904t204546) in Bakersfield, CA, as shown in Figure 2.

We moved the sentence about the background covariance matrix to later in Section 3 (lines 205–208).

In the MF method, the background covariance matrix  $\Sigma$  and mean radiance  $\mu$  are drawn from a reference region close to the CH<sub>4</sub> emission source. These are shown in Figure 2, where the dashed green box denotes the reference region and the source is located within the solid red box.

10. L167, it might be good to describe some of the retrieval artifacts and why they are produced. Are some of the retrieval artifacts related to aerosols or surface albedo?

We have added a description of the artifacts (lines 191–193).

Some artifacts caused by surfaces with strong absorption in the 2100–2500 nm wavelength range, such as oil-based paints or roofs with calcite as a component (Thorpe et al., 2013), also produce large  $\alpha$  values in the MF method

11. L169-172, the first sentence seems to be redundant with previous description and can be removed. Also good to describe how the normalization is done and its main purpose.

We have removed the first sentence. and added some description to explain how and why the normalization is done (lines 197–199).

The normalization is done by calculating the ratio of the radiance to the maximum value across the spectral range, such that the values fall between 0 and 1. This is a first order correction for the effects of surface albedo.

12. L173-174, the sentence “The radiance has units can be removed as the spectral range has already been mentioned earlier in the paragraph and it is not necessary to mention the units of radiance.

We have deleted the sentence.

13. L206, the spectral resolution of 0.5cm<sup>-1</sup> seems to be too coarse to resolve monochromatic spectral features in this spectral region. Have you performed sensitivity calculation to see how this affect the synthetic AVIRIS-NG radiance?



AVIRIS-NG has 400 channels overall, and 80 within the 2100–2500 nm range. The spectral resolution of  $0.5 \text{ cm}^{-1}$  provides more than 1520 monochromatic wavelengths. Therefore, there are around 20 monochromatic calculations per spectral channel, which should be sufficient. Nevertheless, we performed a sensitivity calculation as recommended by the reviewer. Changing the spectral resolution to  $0.1 \text{ cm}^{-1}$  resulted in  $\sim 1\%$  difference in the retrieved  $\text{XCH}_4$ . We have added Figure 9c to show the effect of spectral resolution on the retrieval.

14. L178-186, it is good to mention clearly whether aerosol is included in both the forward model and retrieval. It seems that aerosol is not retrieved, but not sure if fixed aerosol model is used in the forward model as it mentions “Single scattering ... using all moments of the phase function”

For the AVIRIS-NG retrievals, aerosols are neither included in the forward model nor in the retrieval. This is now clearly mentioned in Section 2.2 (lines 172–173).

Aerosols are not included in the state vector for both the real and synthetic retrievals. They are, however, considered in the forward model for the synthetic simulations.

This point is reiterated in Section 3 (line 217).

Aerosols are neither included in the forward model nor retrieved in this analysis.

15. L187-191, although  $\text{H}_2\text{O}$  is not retrieved and taken into account in the MF method, it should also cause retrieval bias/uncertainty to the MF result. Probably it will cause different retrieval errors to MF method and OE method due to its different retrieval treatments. Also are some of the differences due to aerosols and surface albedo?

In the MF method, the background mean radiance and covariance matrix are drawn from a region near the plume. We assume that the region used to compute the background radiance has the same  $\text{H}_2\text{O}$  concentration as the plume. However, in the OE method, we retrieve  $\text{H}_2\text{O}$  and  $\text{CH}_4$  simultaneously. These gases have overlapping absorption in the retrieval wavelengths. Note that we normalize the radiance to reduce the influence of surface albedo. Further, aerosols are not considered in either retrieval. Therefore, it is likely that the difference in the treatment of  $\text{H}_2\text{O}$  is the main source of the discrepancy in the retrievals. We have modified the following sentence to clarify this point (lines 221–224).

Since radiance normalization reduces the impact of surface albedo and aerosols are not included in either retrieval, this might be due to the fact that, in the OE method, H<sub>2</sub>O and CH<sub>4</sub> are simultaneously retrieved; the CH<sub>4</sub> retrieval has added uncertainty due to overlapping absorption features between these two gases.

16. L226, the absorption cross-section is independent of concentration, suggest removing “cross-section”

We have made the suggested change.

17. L228, why does the effect of aerosol loading cause underestimation? Would be good to provide some explanation. Due to the shielding of CH<sub>4</sub> absorption below aerosol layer?

The reviewer is right. The effect of aerosol loading is to shield CH<sub>4</sub> absorption below the aerosol layer. There is also another effect — it increases the path length (due to multiple scattering between the aerosol layer and the surface). When aerosol scattering is ignored in the retrieval, it manifests as reduced CH<sub>4</sub> absorption. We add a brief description per the reviewer’s suggestion (lines 265–267).

The underestimation, which is due to the shielding of CH<sub>4</sub> absorption below the aerosol layer and the fact that multiple scattering effects between the aerosol and the surface are ignored, is clearly shown in Figure 5b

18. L232-233 and in Fig. 5c, are the results really independent of surface albedo here? Or is this simply because a background with the same surface albedo is used? In actual MF retrievals, surface albedo is not necessarily known (or be the same as that in the background). Also according to normalization procedure shown in Fig. 2, looks like most of the surface albedo can be taken into account after the normalization if surface albedo is not perfectly known as in real retrievals, but bias will occur. Please clarify this.

We assume different surface albedos to calculate the covariance matrix and mean background radiance for the MF method. We did calculate retrieval biases for different surface albedos, but the results are indistinguishable. Hence, we just include one line in Figure 5c. The reason for this behavior is that when there is no aerosol loading, there are no multiple scattering effects between the surface and the atmosphere (Rayleigh scattering is negligible in the retrieval wavelength range). We add the following sentence (lines 271–

273) to clarify this point.

This is because there are no multiple scattering effects between the surface and the atmosphere (Rayleigh scattering is negligible in the retrieval wavelength range) when there is no aerosol loading.

The reviewer's point is well taken, though. The results clearly show that the enhancement depends on the surface albedo when  $AOD > 0$ . The spread increases as the AOD becomes larger. We add another sentence (lines 279–280) to point that out.

The implication of these results is that accurate knowledge of the surface albedo is important for MF retrievals, especially when the aerosol loading is large.

19. L236, good to explain why larger biases at large AOD and surface albedo values. Also since the enhancement in units of ppm m is retrieved with the MF method, it is better to mention the bias in enhancement ( $\sim -700$  ppm m) rather than saying “maximum bias .. close to  $0.06 \times 1.822$  ppm), or you can say “the maximum bias is close to  $\sim -700$  ppm m (equivalent of  $-0.06 \times 1.822$  ppm) ...”

We have implemented the reviewer's suggestion and added further description of the physical reasoning behind the larger biases (lines 273–279).

For the scenarios with aerosol loading, the dispersion in the zero-enhancement  $XCH_4$  value between different surface albedos indicates that results from the MF method are biased more at large AOD and surface albedo values (Figures 5c–f). This is a consequence of increased multiple scattering between the aerosol layer and the surface that is not accounted for by the retrieval algorithm. The maximum bias value is close to  $-700$  ppm  $\times$  m (equivalent to  $-0.06 \times 1.822$  ppm relative to the background concentration of  $1.0 \times 1.822$  ppm) for an AOD of 0.3 and albedo of 0.5 (Figure 5f).

20. L242-243, why does the bias decreases with increasing  $CH_4$  concentration for the MF method? The reason gives on L244-245 only shows that the enhancement is more underestimated at larger  $XCH_4$  concentration (as shown from the curves in Figure 5a that

deviated from a straight line), and seems not able to explain the enhancement difference between without and with aerosols decreases with increasing CH<sub>4</sub> concentration.

The reviewer is right. The enhancement underestimation at large XCH<sub>4</sub> values is shown by the deviation of Figure 5a from a straight line. The enhancement bias trend is a result of the changing slope of this curve as a function of XCH<sub>4</sub>, which in turn is due to the increasing inaccuracy of the linear MF method at large XCH<sub>4</sub> values, where the absorption behavior is highly nonlinear. We explain this in lines 288–292.

This surprising behavior is a direct consequence of the physical basis of the MF method. The rate of increase in enhancement becomes smaller as XCH<sub>4</sub> becomes larger (Figure 5a). Therefore, at higher XCH<sub>4</sub> values, the addition of aerosols (which has a similar effect as a reduction in XCH<sub>4</sub>) results in a lower reduction in enhancement compared to that at lower XCH<sub>4</sub> values, resulting in a net decrease in the enhancement bias.

21. Figure 6, the bias is negative (underestimated) as indicated in the text. Suggest making it clear in Fig. 6 caption that the figure shows the magnitude of underestimation.

We apologize for the confusing definition. The bias is defined as the difference between the enhancement values without and with aerosol (i.e. without-with). We clarify this in the revised manuscript (lines 282–284).

The color bar shows the  $\alpha$  bias — which is defined as the difference between the enhancement value without aerosol (true  $\alpha$  value) and that with aerosol — for different CH<sub>4</sub> concentrations, surface albedos and AODs. A positive bias means that CH<sub>4</sub> is underestimated.

22. L255 and Figures 7,8,9, is the bias also negative? If so, please make it clear.

The bias here is defined as the difference between the true and retrieved XCH<sub>4</sub>. Therefore, the bias is positive. We clarify this in the revised manuscript (lines 297–298).

the retrieval bias is defined as the difference between the true XCH<sub>4</sub> in the simulation and the retrieved value (positive values refer to underestimation).

23. L258, good to explain how the bias varies with SSA and g.

We add the following sentences (lines 302–305) to explain the bias variation with SSA and  $g$ .

This behavior can be explained as follows. At higher SSA values, there are more multiple scattering effects (that are ignored in the retrieval). On the other hand, larger values of  $g$  imply greater anisotropy of scattering (preference for forward scattering), leading to a reduction in multiple scattering effects.

24. L259-267 and results in Figure 8: is surface albedo retrieved? It seems to me that it is retrieved with XCH<sub>4</sub> so that the error is small for different surface albedo when AOD=0. While for other cases (e.g., surface albedo is kept constant at 0.3), maybe surface albedo is not retrieved. Please make it clear probably at the end of section 2.2 or in this paragraph about whether surface albedo is retrieved and how it is retrieved (e.g., wavelength independent or dependent)

Please note that simulations for this figure are done for AOD between 0.1 and 1. In any case, for AOD = 0, there are no multiple scattering effects between the aerosol and the surface and hence no uncertainty in path length. This is why the error is small for different surface albedos for this scenario. The surface albedo is not retrieved. In the simulations, we set it to a wavelength-independent value. We describe this in Section 2.2 (lines 178–179) and in Section 4.3 (lines 295–296).

The surface albedo is also not retrieved; for both real and synthetic retrievals, it is held fixed and assumed to be independent of wavelength.

The OE method is then used to perform retrievals using the same configuration (including, in particular, the same surface albedo) except that AOD is set to zero.

25. L264, it seems to me that the bias is defined differently for the MF case, as the enhancement between without and with aerosols (L239-240), while the bias for OE is defined as the difference between retrieved and true XCH<sub>4</sub>. If we use a similar definition, according to Figure 5a, there is larger underestimation at higher XCH<sub>4</sub> values for both without and with aerosols in the MF method.

It is not possible to use similar definitions for the two methods. The OE method retrieves

XCH<sub>4</sub>, while the MF method retrieves the enhancement. Further, Figure 5a shows the enhancement as a function of XCH<sub>4</sub>. The enhancement increases with XCH<sub>4</sub>. That is not the same thing as saying that the XCH<sub>4</sub> biases are larger. The enhancement bias (plotted in Figure 6) is a better representation of the effects of aerosol scattering.

26. L273, please check if it should be between “with and without aerosols” as the case with zero AOD is the truth reference for the MF method.

The reviewer is right. We have rephrased the sentence as follows (lines 337–338).

On the other hand, in the OE method, it is the ratio of the bias to the true XCH<sub>4</sub>.

27. L275-281, how is this OE retrieval sensitive to the assumed a priori error of 20%? If you use a larger a priori error for the OE method, will the conclusion here be changed?

The effect of changing the *a priori* error is described in lines 330–333.

Similarly, the XCH<sub>4</sub> difference is less than 4 ppb when the *a priori* error changes from 0.05 to 0.5 (Figure 9b). Compared to the bias of about 923 ppb induced by neglecting aerosol scattering for this scenario, it is clear that the impacts of the *a priori* and *a priori* error are very small.

28. L280, an example is given for a XCH<sub>4</sub> of 1.1x1.822ppm. It is useful to give another example at high XCH<sub>4</sub>, for example XCH<sub>4</sub>=5.0.

We provide bias ratio comparisons for a range of scenarios (lines 339–341).

The bias ratio for the MF method (1.3–4.5%) is up to 53.6% less than that for the OE method (2.8–5.6%) for AOD = 0.3 when the CH<sub>4</sub> concentration is high (2–5 times typical background values).

29. Section 5 is a summary of this paper and discussion about future work, I suggest changing this section title to “Summary and discussion”

We have made the suggested change.

30. L289-291, the sentence might not be true as mentioned earlier due to different bias definitions used for OE and MF methods.

The bias definitions are different by necessity, as described in the response to comment #25. Within the framework of those definitions, the statement holds.