

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 provides some analysis about how aerosols properties affect CH₄ retrieval, which will attract a lot of interests from the audience of this journal. However, it is suggested that more specific analysis about the aerosol model are needed and the main points about aerosol impact need to be emphasized in both abstract and main part.

We thank the reviewer for the excellent suggestion. We have added some sentences (lines 27–30) in the abstract.

The presence of aerosols causes an underestimation of CH₄ 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.

We added Tables 1 and 2 and provided a description of the aerosol models (lines 173–178).

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).

We also added more description of the aerosol impact in the main text (lines 305–312). Further, we added Figures 7b and 7c.

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.

Moreover, in the two retrieval algorithm used in this study, no aerosol loading is included. I'm just wondering if AOD or other aerosol parameters are retrieved simultaneously with XCH₄, such as adding AOD in the state vector of OE retrieval, will the retrieval bias be improved? If any preliminary results could be shown, it will be interesting.

We appreciate the reviewer's suggestion. The issue is that the MF method does not permit retrieval of AOD; it has traditionally been intended to provide a quick detection of CH₄. The OE method, on the other hand, is more flexible and does allow aerosol retrieval. We did not add AOD to the state vector since one of the methods was incapable of handling it, and we would not be able to make a meaningful comparison. In this work, we instead study the aerosol impact indirectly, by including it in the simulations but not in the retrieval. Through this process, we demonstrate that the MF method has larger biases for diffuse sources. We indicate in the abstract (lines 24–27) that the AOD is not included in the state vector.

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 reviewer makes an important point, though. We modify/add the following sentences (lines 344–351) indicating that the MF method is also not optimal for scenarios with aerosol scattering.

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 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 CH₄ concentration.

We also add two sentences (lines 37–40) in the abstract and add/modify some sentences (lines 362–364, 369–371) in the summary.

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 CH₄. The results indicate that, while the MF method is good for plume detection, the OE method should be employed to quantify CH₄ concentrations, especially in the presence of aerosol scattering.

The MF method shows smaller bias ratios at large CH₄ concentrations than the OE method; it is, therefore, the optimal method to detect strong CH₄ emission sources when scattering effects can be ignored in the retrieval.

Therefore, when scattering effects need to be considered, the OE method is the appropriate choice. Indeed, the MF method was intended for plume detection. OE enables accurate quantification of XCH₄ in the presence of aerosol scattering.

Furthermore, the section 3 has less close relationship with the topic of this paper, the authors are suggested to think it more.

We feel that this section belongs in the paper. Section 3 provides a comparison of MF and OE retrievals from a real AVIRIS-NG measurement. These results provide heuristic information about the relative performance of the two techniques. However, there are some difficulties in comparing these retrievals. Further, understanding the retrieval effects of ignoring aerosol scattering is easier when we employ simulations. Therefore, both methods of comparison are useful and illustrative. We add/modify the following sentences (lines 225–231):

While these results provide heuristic information about the relative performance of the two retrieval techniques, it is difficult to compare the CH₄ enhancement directly between the two methods since the background CH₄ concentration used in the MF method cannot be quantified exactly. Further, evaluating retrieval biases due to ignoring aerosol scattering is not trivial when real measurements are used. Therefore, we simulate synthetic spectra (see section 4) using the 2S-ESS RT model to study the impacts of aerosol scattering as a function of different geophysical parameters by varying them in a systematic manner.

Specific comments

1. In the third paragraph of Introduction, I suggest the authors to add more the description about how to retrieve CH₄ concentration from satellite measurements, especially the advantage of hyperspectral imaging in CH₄ retrieval. I think the description about atmospheric correction has less relationship with the topic of this paper.

The reviewer is right. The description of atmospheric correction did not flow well with the rest of the introduction. We removed that paragraph and added a paragraph on satellite retrieval of CH₄ concentrations (lines 68–84).

Satellite monitoring of CH₄ can be broadly divided into three categories: solar backscatter, thermal emission and lidar (Jacob et al., 2016). The first solar backscattering mission was SCIAMACHY (Frankenberg et al., 2006), which was operational from 2003–2012 and observed the entire planet once every seven days. It was followed by GOSAT in 2009 (Kuze et al., 2016), and subsequently the next generation GOSAT-2 in 2018 (Glumb et al., 2014). In between, the TROPOMI mission was also launched in 2017, which observes the planet once daily with a high spatial resolution of 7×7 km² (Butz et al., 2012; Veefkind et al., 2012). CarbonSat (Buchwitz et al., 2013) is another proposed mission to measure CH₄ globally from solar backscatter with a very fine spatial resolution (2×2 km²) and high precision (0.4%). Thermal infrared observations of CH₄ are available from the IMG (Clerbaux et al., 2003), AIRS (Xiong et al., 2008), TES (Worden et al., 2012), IASI (Xiong et al., 2013), and CrIS (Gambacorta et al., 2016) instruments. These instruments provide day/night measurements at spatial resolutions ranging from 5×8 km² (TES) to 45×45 km² (AIRS). GEO-CAPE (Fishman et al., 2012), GeoFTS (Xi et al., 2015), G3E (Butz et al., 2015), and GeoCarb (Polonsky et al., 2014) are proposed geostationary instruments (GeoCarb was selected by NASA under the Earth Venture - Mission program), which when operational will have resolutions of 2–5 km over regional scales. The MERLIN lidar instrument (Kiemle et al., 2014) scheduled for launch in 2021 will measure CH₄ by employing a differential absorption lidar.

2. Line 171-172: How to do normalization for measured radiance? Add some description about this, please.

We have 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.

3. Line 181: Is the typical XCH₄ background of 1.822 ppm shown by the authors here related to the background covariance matrix and mean radiance used in MF method? Some reasons are expected here. By the way, it is better to mention the background covariance matrix and mean radiance in MF retrieval of CH₄ plume case here.

For a real AVIRIS-NG measurement, the mean radiance and background covariance matrix used in the MF method are taken from a reference region close to the CH₄ plume source. For the OE method in this case, a typical XCH₄ background of 1.822 ppm is used. This typical value is obtained from annual mean data tabulated by the NOAA Global Monitoring Laboratory. For synthetic MF retrievals, we compute the mean radiance and background covariance matrix using simulations for the same typical XCH₄ background of 1.822 ppm at different values of the surface albedo. We indicate how we obtained the typical value for the OE model in lines 208–211.

In the OE method, results are shown as a multiplicative scaling factor compared to a typical XCH₄ background of 1.822 ppm. This value is the globally averaged marine surface annual mean for 2014 (Ed Dlugokencky, NOAA/GML (www.esrl.noaa.gov/gmd/ccgg/trends_ch4/), the year corresponding to the AVIRIS-NG measurement being studied).

4. In the OE retrieval in section 3, what is the definition of the a priori value of XCH₄? What aerosol model do the authors use? I think some parameters about aerosol model are expected here.

We have added the following description (lines 170–178) to provide the information the reviewer requested:

The *a priori* values are within 10% of the true values; *a priori* errors are assumed to be 20% for all state vector elements. The retrieved results are shown as the column averaged mixing ratio (X_{CH_4} , in ppm). 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).

5. In section 4.3, the authors show the variation of OE X_{CH_4} retrieval bias with SSA, g , AOD, surface albedo and X_{CH_4} . Which parameters affect X_{CH_4} retrieval bias most? From aerosol parameters, which type of aerosols, such as smoke, dust or sea salt, causes the largest or lowest bias in X_{CH_4} retrieval? These information will attract the audience's interest and provide guidance to correct aerosol impact in future X_{CH_4} retrieval algorithm.

We have added the following sentences (lines 302–312) to describe the effects of SSA and g on the retrieval. We also added Figures 7b and 7c.

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 AOD and surface albedo impacts are compared in lines 321–324.

The CH₄ bias induced by differences in the surface albedo is not as large as that due to AOD variations, but albedo effects are noticeable at large AOD. Figure 8d shows the sensitivity of retrieval biases to changes in AOD and surface albedo, again demonstrating the greater impact of AOD than surface albedo in the retrieval.

6. In OE retrieval, the *a priori* error of XCH₄ will affect the retrieval bias as well. Maybe the authors could check its impact.

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

Similarly, the XCH₄ 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.

Technical corrections

1. Figure 9a and 9b have some overlaps with the same XCH₄. There is no need to express them using two figures.

Our objective is to show two regimes, one where the OE method has lower bias ratio and the other where the MF method performs better. There is a crossover region between ~1.5–2 where both methods produce similar results. We believe that using two figures shows this behavior clearly. Some of the details might be lost if they were combined into one.

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9. Explanations are supplied for the behavior of the retrievals as a function of the different parameters.
10. More quantification is provided for the differences between the two retrieval techniques.
11. Effects of changing the *a priori*, *a priori* error and simulation spectral resolution are described.
12. 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, CH₄ 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 CH₄ 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 CH₄ concentrations (2–5 times typical background values), and is suitable for detecting strong CH₄ 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 CH₄. The results indicate that, while the MF method is good for plume detection, the OE method should be employed to quantify CH₄ 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 XCH₄ 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 $XCH_4 = 5.8 \times 1.822$ 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_4 changes by about 9 ppb as the *a priori* changes from half to twice the true XCH_4 value. Similarly, the XCH_4 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_4 still changes by only about 100 ppb when the spectral resolution is changed from 0.5 to 0.1 cm^{-1} (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_4 concentration is high (2–5 times typical background values). On the other hand, the OE method performs better when enhancements are small and XCH_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 ($XCH_4 = 1.1 \times 1.822$ 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. Therefore, the OE technique is in general superior due to its ability to support simultaneous retrieval of aerosols, surface albedo and CH_4 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 CH₄ 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 CH₄ 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 CH₄ 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 (~ 2-6% for OE and 1,5-3.5% 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 CH₄ concentrations (2–5 times typical background values), and is suitable for detecting strong CH₄ 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 CH₄. The results indicate that, while the MF method is good for plume detection, the OE method should be employed to quantify CH₄ 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 CH₄ concentration is high (2–5 times typical background values). On the other hand, the OE method performs better when enhancements are small and XCH₄ 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 (XCH₄ = 1.1 × 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 CH₄ 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 CH₄ 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 CH₄ concentrations (2–5 times typical background values), and is suitable for detecting strong CH₄ 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 CH₄ 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 $10^{-3} \text{ m}^3/\text{mol}$, and $1/(V \cdot 10^3)$ has a unit of mol/m^3 . Maybe $1/(V \cdot 10^3)$ should be $V \cdot 10^{-3}$ 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):

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

κ_{trad} 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 μ 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 Σ and mean radiance μ are drawn from a reference region close to the CH_4 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 α 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^{-1} 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 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 cm^{-1} resulted in $\sim 1\%$ difference in the retrieved 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 H_2O 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 H₂O concentration as the plume. However, in the OE method, we retrieve H₂O and CH₄ 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 H₂O 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, H₂O and CH₄ are simultaneously retrieved; the CH₄ 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 CH₄ absorption below aerosol layer?

The reviewer is right. The effect of aerosol loading is to shield CH₄ 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 CH₄ absorption. We add a brief description per the reviewer's suggestion (lines 265–267).

The underestimation, which is due to the shielding of CH₄ 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 (~ -700 ppm m) rather than saying “maximum bias .. close to 0.06×1.822 ppm), or you can say “the maximum bias is close to ~ -700 ppm m (equivalent of -0.06×1.822 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 XCH₄ 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 -0.06×1.822 ppm relative to the background concentration of 1.0×1.822 ppm) for an AOD of 0.3 and albedo of 0.5 (Figure 5f).

20. L242–243, why does the bias decreases with increasing CH₄ concentration for the MF method? The reason gives on L244–245 only shows that the enhancement is more underestimated at larger XCH₄ 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 CH₄ concentration.

The reviewer is right. The enhancement underestimation at large XCH_4 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_4 , which in turn is due to the increasing inaccuracy of the linear MF method at large XCH_4 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_4 becomes larger (Figure 5a). Therefore, at higher XCH_4 values, the addition of aerosols (which has a similar effect as a reduction in XCH_4) results in a lower reduction in enhancement compared to that at lower XCH_4 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 α bias — which is defined as the difference between the enhancement value without aerosol (true α value) and that with aerosol — for different CH_4 concentrations, surface albedos and AODs. A positive bias means that CH_4 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_4 . 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 XCH₄ 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 XCH₄ 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 XCH₄. If we use a similar definition, according to Figure 5a, there is larger underestimation at higher XCH₄ 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 XCH₄, while the MF method retrieves the enhancement. Further, Figure 5a shows the enhancement as a function of XCH₄. The enhancement increases with XCH₄. That is not the same thing as saying that the XCH₄ 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 XCH₄.

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 XCH₄ 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 XCH₄ of 1.1x1.822ppm. It is useful to give another example at high XCH₄, for example XCH₄=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 CH₄ 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.

1 Quantifying the impact of aerosol scattering on the 2 retrieval of methane from airborne remote sensing 3 measurements

4
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17
18 **Abstract.** As a greenhouse gas with strong global warming potential, atmospheric methane (CH₄)
19 emissions have attracted a great deal of attention. Although remote sensing measurements can provide
20 information about CH₄ sources and emissions, accurate retrieval is challenging due to the influence of
21 atmospheric aerosol scattering. In this study, imaging spectroscopic measurements from the Airborne
22 Visible/Infrared Imaging Spectrometer–Next Generation (AVIRIS-NG) in the short-wave infrared are
23 used to compare two retrieval techniques — the traditional Matched Filter (MF) method and the Optimal
24 Estimation (OE) method, which is a popular approach for trace gas retrievals. Using a numerically
25 efficient two-stream-exact-single-scattering radiative transfer model, we also simulate AVIRIS-NG
26 measurements for different scenarios and quantify the impact of aerosol scattering in the two retrieval
27 schemes by including aerosols in the simulations but not in the retrievals. The presence of aerosols causes
28 an underestimation of CH₄ in both the MF and OE retrievals; the biases increase with increasing surface
29 albedo and aerosol optical depth (AOD). Aerosol types with high single scattering albedo and low
30 asymmetry parameter (such as water soluble aerosols) induce large biases in the retrieval. When
31 scattering effects are neglected, the MF method exhibits lower fractional retrieval bias compared to the
32 OE method at high CH₄ concentrations (2–5 times typical background values), and is suitable for
33 detecting strong CH₄ emissions. For an AOD value of 0.3, the fractional biases of the MF retrievals are

34 between 1.3 and 4.5%, while the corresponding values for OE retrievals are in the 2.8–5.6% range. On
35 the other hand, the OE method is an optimal technique for diffuse sources (<1.5 times typical background
36 values), showing up to five times smaller fractional retrieval bias (8.6%) than the MF method (42.6%)
37 for the same AOD scenario. However, when aerosol scattering is significant, the OE method is superior
38 since it provides a means to reduce biases by simultaneously retrieving AOD, surface albedo and CH₄.
39 The results indicate that, while the MF method is good for plume detection, the OE method should be
40 employed to quantify CH₄ concentrations, especially in the presence of aerosol scattering.

41 1 Introduction

42 Atmospheric methane (CH₄) is about 85 times more potent per unit mass at warming the Earth than
43 carbon dioxide (CO₂) on a 20-year timescale (IPCC, 2013), implying that reduction in CH₄ emissions
44 could be very efficient to slow down global warming in the near term. Global mean CH₄ concentrations
45 have increased from ~700 ppb in the preindustrial era to more than 1860 ppb as of 2019 (NOAA, 2019).
46 The most effective sink of atmospheric CH₄ is the hydroxyl radical (OH) in the troposphere. CH₄ reacts
47 with OH to reduce the oxidizing capacity of the atmosphere and generate tropospheric ozone. Increasing
48 emissions of CH₄ reduce the concentration of OH in the atmosphere. With less OH to react with, the
49 lifespan of CH₄ could also increase, resulting in greater CH₄ concentrations (Holmes et al., 2013). Soils
50 also act as a major sink for atmospheric methane through the methanotrophic bacteria that reside within
51 them.

52 Significant natural CH₄ sources include wetlands (Bubier et al., 1994, Macdonald et al., 1998;
53 Gedney et al., 2004), geological seeps (Kvenvolden and Rogers, 2005; Etiope et al., 2009), ruminant
54 animals, and termites. In addition, increased surface and ocean temperatures associated with global
55 warming may increase CH₄ emissions from melting permafrost (Woodwell et al., 1998; Walter et al.,
56 2006; Schaefer et al., 2014, Schuur et al., 2015) and methane hydrate destabilization (Kvenvolden, 1988;
57 Archer, 2007). Human activity also contributes significantly to the total CH₄ emissions. Rice agriculture
58 is one of the most important anthropogenic sources of CH₄ (Herrero et al., 2016; Schaefer et al., 2016).
59 Other sources include landfills (Themelis and Ulloa, 2007), wastewater treatment, biomass burning, and
60 methane slip from gas engines. Global fugitive CH₄ emissions from coal mining (Kort et al., 2014),
61 natural gas and oil systems (Alvarez et al., 2018), hydraulic fracturing (“fracking”) of shale gas wells
62 (Howarth et al., 2011; Howarth, 2015, 2019), and residential and commercial natural gas distribution
63 sectors (He et al., 2019) are also of increasing concern. Although the sources and sinks of methane are
64 reasonably well known, there are large uncertainties in their relative amounts and in the partitioning
65 between natural and anthropogenic contributions (Nisbet et al., 2014, 2016). This uncertainty is
66 exemplified by the CH₄ “hiatus”, which refers to the observed stabilization of atmospheric CH₄
67 concentrations from 1999–2006, and the renewed rise thereafter (Kirschke et al., 2013).

68 Satellite monitoring of CH₄ can be broadly divided into three categories: solar backscatter, thermal
69 emission and lidar (Jacob et al., 2016). The first solar backscattering mission was SCIAMACHY
70 (Frankenberg et al., 2006), which was operational from 2003–2012 and observed the entire planet once
71 every seven days. It was followed by GOSAT in 2009 (Kuze et al., 2016), and subsequently the next
72 generation GOSAT-2 in 2018 (Glumb et al., 2014). In between, the TROPOMI mission was also
73 launched in 2017, which observes the planet once daily with a high spatial resolution of 7×7 km² (Butz

74 et al., 2012; Veeffkind et al., 2012). CarbonSat (Buchwitz et al., 2013) is another proposed mission to
75 measure CH₄ globally from solar backscatter with a very fine spatial resolution (2×2 km²) and high
76 precision (0.4%). Thermal infrared observations of CH₄ are available from the IMG (Clerbaux et al.,
77 2003), AIRS (Xiong et al., 2008), TES (Worden et al., 2012), IASI (Xiong et al., 2013), and CrIS
78 (Gambacorta et al., 2016) instruments. These instruments provide day/night measurements at spatial
79 resolutions ranging from 5×8 km² (TES) to 45×45 km² (AIRS). GEO-CAPE (Fishman et al., 2012),
80 GeoFTS (Xi et al., 2015), G3E (Butz et al., 2015), and GeoCarb (Polonsky et al., 2014) are proposed
81 geostationary instruments (GeoCarb was selected by NASA under the Earth Venture - Mission program),
82 which when operational will have resolutions of 2–5 km over regional scales. The MERLIN lidar
83 instrument (Kiemle et al., 2014) scheduled for launch in 2021 will measure CH₄ by employing a
84 differential absorption lidar.

85 By combining a large number of footprints and high spatial resolution, airborne imaging
86 spectrometers are also well suited for mapping local CH₄ plumes. The Airborne Visible/Infrared Imaging
87 Spectrometer–Next Generation (AVIRIS-NG) measures reflected solar radiance across more than 400
88 channels between 380 and 2500 nm (Green et al., 1998; Thompson et al., 2015). Strong CH₄ absorption
89 features present between 2100 and 2500 nm can be observed at a spectral resolution of 5 nm full width
90 at half maximum (FWHM). A number of approaches have been developed to retrieve CH₄ from such
91 hyperspectral data. Roberts et al. (2010) used a spectral residual approach between 2000 and 2500 nm
92 and Bradley et al. (2011) employed a band ratio technique using the 2298 nm CH₄ absorption band and
93 2058 nm CO₂ absorption band. However, these techniques are not suited for terrestrial locations that
94 have lower albedos and have spectral structure in the SWIR. A cluster-tuned matched filter technique
95 was demonstrated to be capable of mapping CH₄ plumes from marine and terrestrial sources (Thorpe et
96 al., 2013) as well as CO₂ from power plants (Dennison et al., 2013); however, this method does not
97 directly quantify gas concentrations. Frankenberg et al. (2005) developed an iterative maximum *a*
98 *posteriori* differential optical absorption spectroscopy (IMAP-DOAS) algorithm that allows for
99 uncertainty estimation. Thorpe et al. (2014) adapted the IMAP-DOAS algorithm for gas detection in
100 AVIRIS imagery. In addition, they developed a hybrid approach using singular value decomposition and
101 IMAP-DOAS as a complementary method of quantifying gas concentrations within complex AVIRIS
102 scenes.

103 Accurate assessment of CH₄ emissions is particularly challenging in the presence of aerosols
104 because the latter introduce uncertainties in the light path if not accounted for. In fact, CH₄ emissions are
105 frequently correlated with pollution due to concurrent aerosol emissions. For large aerosols (such as dust),
106 the low Ångström exponent values result in high aerosol optical depth (AOD) values even in the

107 wavelength range from 2000 nm to 2500 nm (Seinfeld and Pandis, 2006; Zhang et al., 2015). Therefore,
 108 it is important to obtain a clear understanding of aerosol impacts on CH₄ retrievals. In this study, SWIR
 109 AVIRIS-NG measurements are used to analyze the impact of aerosol scattering on CH₄ retrievals.
 110 Further, using an accurate but numerically efficient radiative transfer (RT) model (Spurr and Natraj,
 111 2011), we simulate AVIRIS-NG measurements with varying aerosol amounts and quantify the impact of
 112 aerosol scattering using two retrieval techniques, the traditional matched filter (MF) method and the
 113 optimal estimation (OE) method that is widely used in trace gas remote sensing. This article is organized
 114 as follows. The MF and OE retrieval methods are described in Section 2. Section 3 focuses on analysis
 115 of a sample CH₄ plume detected by AVIRIS-NG measurements and compares retrievals using the MF
 116 and OE methods. Section 4 presents a detailed evaluation of aerosol impacts on the two retrieval methods
 117 through simulations of AVIRIS-NG spectra for different geophysical parameters. Section 5 provides a
 118 summary of the work and discusses future research.

119

120 2 Methods

121 2.1 MF method

122 Real-time remote detection using AVIRIS-NG measurements are traditionally based on the MF
 123 method (Frankenberg et al., 2016). In this method, the background spectra are assumed to be distributed
 124 as a multivariate Gaussian \mathcal{N} with covariance matrix Σ and background mean radiance μ . If H_0 is a
 125 scenario without CH₄ enhancement and H_1 is one with CH₄ enhancement, the MF approach is equivalent
 126 to a hypothesis test between the two scenarios:

$$127 \quad H_0: L_m \sim \mathcal{N}(\mu, \Sigma) \quad (1)$$

$$128 \quad H_1: L_m \sim \mathcal{N}(\mu + t\alpha, \Sigma) \quad (2)$$

129 where L_m is the measurement radiance; t is the target signature, which is defined in Equation (4); α is the
 130 enhancement value, denoting a scaling factor for the target signature that perturbs the background μ . If
 131 x is a vector of measurement spectra with one element per wavelength, $\alpha(x)$ can be written, based on
 132 maximum likelihood estimates (Manolakis et al., 2014), as follows:

$$133 \quad \alpha(x) = \frac{(x - \mu)^T \Sigma^{-1} t}{t^T \Sigma^{-1} t} \quad (3)$$

134 We utilize the same definitions as in Frankenberg et al. (2016). Specifically, the enhancement value $\alpha(x)$
 135 denotes the thickness and concentration within a volume of equivalent absorption, and has units of ppm
 136 \times m. The target signature t refers to the derivative of the change in measured radiance with respect to a
 137 change in absorption path length due to an optically thin absorbing layer of CH₄. Note that this definition

138 has the disadvantage that the accuracy of the result degrades when the absorption is strong and further
 139 attenuation becomes nonlinear. At a particular wavelength λ , t can be expressed as:

$$140 \quad t(\lambda) = -\kappa(\lambda)\mu(\lambda), \quad (4)$$

141 where κ is the absorption coefficient for a near-surface plume with units of $\text{ppm}^{-1} \text{m}^{-1}$. This is
 142 different from the units of $\text{m}^2 \cdot \text{mol}^{-1}$ traditionally used for the absorption coefficient κ_{trad} in trace
 143 gas remote sensing. Using the ideal gas law to express the volume V (in liters) occupied by one mole of
 144 CH_4 at the temperature and pressure corresponding to the plume altitude ($V = 22.4$ at standard
 145 temperature and pressure), and the relations $1 \text{ liter} = 10^{-3} \text{ m}^3$ and $1 \text{ ppm} = 10^{-6}$, we obtain the
 146 following expression for unit conversion (units in parentheses):

$$147 \quad \kappa_{trad} [\text{m}^2 \cdot \text{mol}^{-1}] = \kappa [\text{ppm}^{-1} \text{m}^{-1}] \times V [\text{liter mol}^{-1}] \times 10^{-3} [\text{m}^3 \text{ liter}^{-1}] / 10^{-6} [\text{ppm}^{-1}] \quad (5)$$

148 Figure 1 shows the target signature, which is calculated based on HITRAN absorption cross-sections
 149 (Rothman et al., 2009). The background mean radiance μ used in Equation 4 is based on the AVIRIS-
 150 NG measurement shown in Figure 2; this is described in more detail in Section 3.

151 2.2 OE method

152 The OE method is widely used for the remote sensing retrieval of satellite measurements, such as
 153 from the Orbiting Carbon Observatory-2 (OCO-2; O'Dell et al., 2018), the Spinning Enhanced Visible
 154 and Infra-Red Imager (SEVIRI; Merchant et al., 2013), and the Greenhouse Gases Observing Satellite
 155 (GOSAT; Yoshida et al., 2013). It combines an explicit (typically nonlinear) forward model of the
 156 atmospheric state, a (typically Gaussian) prior probability distribution for the variabilities and a (typically
 157 Gaussian) distribution for the spectral measurement errors. In addition, the Bayesian framework used by
 158 the OE approach allows new information (from measurements) to be combined with existing information
 159 (e.g., from models). In many applications, the forward model is nonlinear, and obtaining the optimal
 160 solution requires iterative techniques such as the Levenberg–Marquardt method (Rodgers, 2000), which
 161 has been routinely applied to study the impacts of measurement parameters on the retrieval process (see,
 162 e.g., Zhang et al., 2015). The iteration in this algorithm follows the below procedure.

$$163 \quad \mathbf{x}_{i+1} = \mathbf{x}_i + [(1 + \gamma)\mathbf{S}_a^{-1} + \mathbf{K}_i^T \mathbf{S}_\epsilon^{-1} \mathbf{K}_i]^{-1} \{ \mathbf{K}_i^T \mathbf{S}_\epsilon^{-1} [\mathbf{y} - \mathbf{F}(\mathbf{x}_i)] - \mathbf{S}_a^{-1} [\mathbf{x}_i - \mathbf{x}_a] \} \quad (6)$$

164 where \mathbf{x} is a state vector of surface and atmospheric properties, \mathbf{S}_a is the *a priori* covariance matrix, \mathbf{S}_ϵ
 165 is the spectral radiance noise covariance matrix, \mathbf{K} is the Jacobian matrix, \mathbf{x}_a is the *a priori* state vector
 166 and γ is a parameter determining the size of each iteration step. The measured spectral radiance is denoted
 167 as \mathbf{y} ; $\mathbf{F}(\mathbf{x})$ is the simulated radiance obtained from the forward model. For the retrieval of CH_4 from
 168 AVIRIS-NG measurements, the state vector includes the total column amounts of CH_4 and H_2O , while
 169 for the retrievals from synthetic spectra, the H_2O concentration is fixed and the state vector only includes
 170 the CH_4 total column. The *a priori* values are within 10% of the true values; *a priori* errors are assumed

171 to be 20% for all state vector elements. The retrieved results are shown as the column averaged mixing
172 ratio (XCH₄, in ppm). Aerosols are not included in the state vector for both the real and synthetic
173 retrievals. They are, however, considered in the forward model for the synthetic simulations. Table 1
174 (WCRP, 1986) lists optical properties for four basic aerosol types (dust, water soluble, oceanic and soot).
175 Table 2 (WCRP, 1986) shows the corresponding properties for three aerosol models that are defined as
176 mixtures of the basic components from Table 1. We employ the Henyey-Greenstein phase function
177 (Henyey and Greenstein, 1941), where aerosol composition is determined by two parameters: single
178 scattering albedo (SSA) and asymmetry parameter (*g*). The surface albedo is also not retrieved; for both
179 real and synthetic retrievals, it is held fixed and assumed to be independent of wavelength.

180

181 3 Detection and retrieval of CH₄ from AVIRIS-NG measurements

182 To illustrate the OE retrieval and its difference from the MF method, we perform retrievals for an
183 AVIRIS-NG measurement made on 4 September 2014 (ang20140904t204546) in Bakersfield, CA, as
184 shown in Figure 2. The location is to the west of the Kern Front Oil field. This detection is a case study
185 from the NASA/ESA CO₂ and MEthane eXperiment (COMEX) campaign in California during June and
186 August/September 2014, which includes airborne *in situ*, airborne non-imaging remote sensing, and
187 ground-based *in situ* instruments to provide a real-time remote detection and measurement for CH₄
188 plumes released from anthropogenic sources. An RGB image of flight data is displayed in Figure 2a; the
189 emission source is a pump jack, as described in Thompson et al. (2015). Figure 2b presents results from
190 the MF method, which shows that the CH₄ plume disperses downwind and has a maximum enhancement
191 value of about 2800 ppm × m. Some artifacts caused by surfaces with strong absorption in the 2100–
192 2500 nm wavelength range, such as oil-based paints or roofs with calcite as a component (Thorpe et al.,
193 2013), also produce large α values in the MF method; these can be removed by an optimization method
194 such as the column-wise MF technique (Thompson et al., 2015).

195 Figure 3 displays the measured radiance (a) before normalization and (b) after normalization,
196 corresponding to two detector elements (in plume and out of plume). Every element is a cross-track
197 spatial location. The normalization is done by calculating the ratio of the radiance to the maximum value
198 across the spectral range, such that the values fall between 0 and 1. This is a first order correction for the
199 effects of surface albedo. Comparing the measured spectrum in plume to that out of plume, there is
200 obvious enhancement of CH₄ that is particularly evident in the normalized radiance. CH₄ is the main
201 absorber in the 2100–2500 nm wavelength range, and H₂O is the major interfering gas. Figure 3b
202 indicates the absorption peaks due to H₂O and CH₄.

203 We choose the plume center with 500 elements to illustrate results obtained using the MF and OE
204 methods. The former evaluates the CH₄ α value compared to the background CH₄ concentration, while
205 the latter retrieves XCH₄. In the MF method, the background covariance matrix Σ and mean radiance
206 μ are drawn from a reference region close to the CH₄ emission source. These are shown in Figure 2,
207 where the dashed green box denotes the reference region and the source is located within the solid red
208 box. In the OE method, results are shown as a multiplicative scaling factor compared to a typical XCH₄
209 background of 1.822 ppm. This value is the globally averaged marine surface annual mean for 2014 (Ed
210 Dlugokencky, NOAA/GML, www.esrl.noaa.gov/gmd/ccgg/trends_ch4/), the year corresponding to the
211 AVIRIS-NG measurement being studied. We use an accurate and numerically efficient two-stream-
212 exact-single-scattering (2S-ESS) RT model (Spurr and Natraj, 2011). This forward model is different
213 from a typical two-stream model in that the two-stream approximation is used only to calculate the
214 contribution of multiple scattering to the radiation field. Single scattering is treated in a numerically exact
215 manner using all moments of the phase function. This model has been used for remote sensing of
216 greenhouse gases and aerosols (Xi et al., 2015; Zhang et al., 2015, 2016; Zeng et al., 2017, 2018).
217 Aerosols are neither included in the forward model nor retrieved in this analysis. The surface albedo is
218 set to a wavelength-independent value of 0.5.

219 Results from the two retrieval methods reveal a similar CH₄ plume shape (Figure 4), especially for
220 elements with high CH₄ enhancement. However, larger differences in CH₄ concentrations are evident in
221 the OE retrievals (Figure 4b). Since radiance normalization reduces the impact of surface albedo and
222 aerosols are not included in either retrieval, this might be due to the fact that, in the OE method, H₂O and
223 CH₄ are simultaneously retrieved; the CH₄ retrieval has added uncertainty due to overlapping absorption
224 features between these two gases. The large maximum value of about 3000 in the MF method also
225 contributes to a reduction in relative contrast. While these results provide heuristic information about the
226 relative performance of the two retrieval techniques, it is difficult to compare the CH₄ enhancement
227 directly between the two methods since the background CH₄ concentration used in the MF method cannot
228 be quantified exactly. Further, evaluating retrieval biases due to ignoring aerosol scattering is not trivial
229 when real measurements are used. Therefore, we simulate synthetic spectra (see section 4) using the 2S-
230 ESS RT model to study the impacts of aerosol scattering as a function of different geophysical parameters
231 by varying them in a systematic manner.

232

233 4 Aerosol impact analysis

234 4.1 Synthetic spectra

235 In a real AVIRIS-NG observation, the exact column concentration of CH₄ cannot be controlled.
236 However, synthetic simulations allow us to manipulate parameters such as CH₄ concentration, surface
237 albedo, AOD, g , and SSA, and thereby test aerosol impacts on CH₄ retrievals. The 2S-ESS RT model is
238 used to simulate the AVIRIS-NG spectral radiance. In this model, a prior atmospheric profile with 70
239 layers from the surface up to 70 km is derived from National Center for Environmental Prediction
240 reanalysis data (Kalnay et al., 1996); absorption coefficients for all relevant gases are obtained from the
241 HITRAN database (Rothman et al., 2009). Monochromatic RT calculations are performed at a spectral
242 resolution of 0.5 cm⁻¹; the radiance spectrum is then convolved using a Gaussian instrument line shape
243 function with a wavelength-dependent full width at half maximum (FWHM) from a calibrated AVIRIS-
244 NG data file. The signal to noise ratio (SNR) is set to be 300, with Gaussian white noise added. This
245 procedure results in a wavelength grid with a resolution of about 5 nm. The spectral wavelength range
246 used to retrieve CH₄ is from 2100 nm to 2500 nm.

247 The additional atmospheric and geometric variables included in the model are listed in Table 3,
248 which are held constant unless otherwise mentioned. The observation geometry parameters are taken
249 from a real AVIRIS-NG measurement. Recent AVIRIS-NG flight campaigns have sensor heights ranging
250 from 0.43 to 3.8 km; we choose a value of 1 km, the same as the highest level where aerosol is present
251 in our simulations. The influence of AOD on CH₄ retrieval as a function of SSA and g is analyzed in
252 Section 4.3; in all other cases, SSA and g are held constant at 0.95 and 0.75, respectively, which is
253 representative of aerosols in the Los Angeles region (Zhang et al., 2015).

254 4.2 Aerosol impact in the MF method

255 We simulate synthetic spectra at different AOD, surface albedo and CH₄ concentration values, use
256 the MF method to obtain the CH₄ enhancement, and compare differences in α between scenarios without
257 and with aerosol. The covariance matrix and background mean radiance are calculated from a simulated
258 zero AOD background with surface albedos from 0.1 to 0.5, and XCH₄ set at the typical background
259 value of 1.822 ppm used in Section 3. Figure 5a shows the enhancement value as a function of XCH₄.
260 As the CH₄ concentration increases, the enhancement value obtained by the MF method at first increases
261 approximately linearly. However, the absorption changes in a nonlinear fashion with concentration,
262 whereas the MF method applies a linear formalism to the change. Therefore, the enhancement value
263 (which is correlated with the absorption signature) also shows a deviation from linear behavior at larger
264 XCH₄. Two aerosol scenarios (AOD = 0, 0.3) are compared in Figure 5a, which reveals that the effect of
265 aerosol loading is similar to an underestimation of CH₄ in the retrieval. The underestimation, which is
266 due to the shielding of CH₄ absorption below the aerosol layer and the fact that multiple scattering effects
267 between the aerosol and the surface are ignored, is clearly shown in Figure 5b, where the enhancement

268 value for fixed CH₄ concentration (same concentration as the background) decreases from 0 ppm × m
269 to −1532 ppm × m with increasing AOD. To clarify the impact of AOD at different surface albedo values,
270 zoomed in versions of α as a function of XCH₄ are presented in Figures 5c–f. For the AOD = 0 scenario,
271 the results are independent of surface albedo. This is because there are no multiple scattering effects
272 between the surface and the atmosphere (Rayleigh scattering is negligible in the retrieval wavelength
273 range) when there is no aerosol loading. For the scenarios with aerosol loading, the dispersion in the
274 zero-enhancement XCH₄ value between different surface albedos indicates that results from the MF
275 method are biased more at large AOD and surface albedo values (Figures 5d–f). This is a consequence
276 of increased multiple scattering between the aerosol layer and the surface that is not accounted for by the
277 retrieval algorithm. The maximum bias value is close to −700 ppm × m (equivalent to -0.06×1.822
278 ppm relative to the background concentration of 1.0×1.822 ppm) for an AOD of 0.3 and surface albedo
279 of 0.5 (Figure 5f). The implication of these results is that accurate knowledge of the surface albedo is
280 important for MF retrievals, especially when the aerosol loading is large.

281 A quantitative analysis of underestimation of CH₄ concentration due to aerosol scattering is
282 presented in Figure 6. The color bar shows the α bias — which is defined as the difference between the
283 enhancement value without aerosol (true α value) and that with aerosol — for different CH₄
284 concentrations, surface albedos and AODs. A positive bias means that CH₄ is underestimated. The α bias
285 increases with increasing surface albedo and AOD, reaching a maximum value of about 700 ppm × m
286 for the simulated cases. However, it is interesting that the bias decreases with increasing CH₄
287 concentration, which is different from the results obtained by the OE method (discussed in Section 4.3).
288 This surprising behavior is a direct consequence of the physical basis of the MF method. The rate of
289 increase in enhancement becomes smaller as XCH₄ becomes larger (Figure 5a). Therefore, at higher
290 XCH₄ values, the addition of aerosols (which has a similar effect as a reduction in XCH₄) results in a
291 lower reduction in enhancement compared to that at lower XCH₄ values, resulting in a net decrease in
292 the enhancement bias.

293 4.3 Aerosol impact in the OE method

294 For the simulation of the synthetic spectra, we assume nonzero aerosol loading below 1 km elevation.
295 The OE method is then used to perform retrievals using the same configuration (including, in particular,
296 the same surface albedo) except that AOD is set to zero. This approach is similar to neglecting aerosol
297 scattering in the CH₄ retrieval; the retrieval bias is defined as the difference between the true XCH₄ in
298 the simulation and the retrieved value (positive values refer to underestimation). First, we study the
299 retrieval bias caused by different aerosol types and mixtures. Figure 7a shows CH₄ retrieval biases as a
300 function of SSA and g ; surface albedo and AOD are kept constant at 0.3 and XCH₄ is assumed to be 1.0

301 $\times 1.822$ ppm. The retrieval bias increases with SSA and decreases with g , with a maximum bias ratio
302 (ratio of retrieval bias to the true value) of about 20%. This behavior can be explained as follows. At
303 higher SSA values, there are more multiple scattering effects (that are ignored in the retrieval). On the
304 other hand, larger values of g imply greater anisotropy of scattering (preference for forward scattering),
305 leading to a reduction in multiple scattering effects. Since the retrieval bias is large for high SSA and
306 low g , the water-soluble aerosol type (Table 1) and the maritime aerosol model (Table 2) can be expected
307 to induce greater biases in the retrieval. In order to compare the impacts of SSA and g in further detail,
308 retrieval results due to a $\pm 5\%$ change in SSA and g for the three aerosol models from Table 2 are shown
309 in Figures 7b and 7c. Note that for the maritime aerosol model, the SSA is set to 0.999 for the +5%
310 scenario to ensure physicality. It is clear that (1) the maritime aerosol model induces larger retrieval
311 biases than the other aerosol types, and (2) the retrieval results are more sensitive to changes in g than
312 those in SSA.

313 We then simulate synthetic spectra for different values of CH_4 concentration, surface albedo and
314 AOD. The impacts of aerosol scattering on the retrievals for these scenarios are demonstrated in Figure
315 8. Figure 8a shows a 5×5 panel of boxes. Within each box, XCH_4 is constant, while surface albedo
316 increases from top to bottom and AOD increases from left to right. The variation of XCH_4 across the
317 boxes is shown in Figure 8b. We also show a zoomed in plot of the bottom right box ($\text{XCH}_4 = 5.8 \times$
318 1.822 ppm) in Figure 8c, which illustrates the AOD and surface albedo changes within a box. These
319 changes are identical for all boxes. Figure 8a indicates that OE retrievals produce larger CH_4 biases at
320 higher XCH_4 values, in contrast with MF results. In addition, it is evident that the retrieved CH_4 bias
321 increases with increasing AOD. The CH_4 bias induced by differences in the surface albedo is not as large
322 as that due to AOD variations, but surface albedo effects are noticeable at large AOD. Figure 8d shows
323 the sensitivity of retrieval biases to changes in AOD and surface albedo, again demonstrating the greater
324 impact of AOD than surface albedo in the retrieval.

325 The effects of changing the *a priori*, *a priori* error and RT simulation spectral resolution on the
326 retrieved XCH_4 are shown in Figure 9. For these calculations, the other parameters are set as follows:
327 $\text{SSA} = 0.95$, $g = 0.75$, $\text{AOD} = 1.0$, surface albedo = 0.5 and true $\text{XCH}_4 = 5.8 \times 1.822$ ppm. The parameters
328 were chosen to correspond to the scenario with the largest retrieval bias in Figure 8c (bottom right box
329 in Figure 8c). Figure 9a shows that the retrieved XCH_4 changes by about 9 ppb as the *a priori* changes
330 from half to twice the true XCH_4 value. Similarly, the XCH_4 difference is less than 4 ppb when the *a*
331 *priori* error changes from 0.05 to 0.5 (Figure 9b). Compared to the bias of about 923 ppb induced by
332 neglecting aerosol scattering for this scenario, it is clear that the impacts of the *a priori* and *a priori* error

333 are very small. The effect of spectral resolution is larger, but XCH₄ still changes by only about 100 ppb
334 when the spectral resolution is changed from 0.5 to 0.1 cm⁻¹ (Figure 9c).

335 4.4 Comparison of the two retrieval techniques

336 Figure 10 presents the bias ratios for the two retrieval techniques at different AODs (surface albedo
337 = 0.3). In the MF method, the bias ratio is defined as the ratio of the bias to the true value of α . On the
338 other hand, in the OE method, it is the ratio of the bias to the true XCH₄. From Figure 10 it is clear that
339 the bias ratio decreases with increasing CH₄ concentration and has higher values at larger AODs. The
340 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
341 AOD = 0.3 when the CH₄ concentration is high (2–5 times typical background values). On the other
342 hand, the OE method performs better when enhancements are small and XCH₄ is close to the background
343 value. For example, the bias ratio for the MF method has a high value of about 42.6% at AOD = 0.3 for
344 a 10% enhancement (XCH₄ = 1.1 × 1.822 ppm); the OE value for the same scenario is 8.6%. For scenarios
345 where scattering is ignored, the two retrieval techniques seem to be complementary, with differing
346 utilities for different enhancements. On the other hand, when RT models that account for scattering
347 effects are employed, the MF technique is suboptimal. Further, MF retrievals rely on accurate
348 characterization of the surface albedo, especially when the aerosol loading is large. Finally, the MF
349 method does not retrieve concentrations, which are necessary to infer fluxes. Therefore, the OE technique
350 is in general superior due to its ability to support simultaneous retrieval of aerosols, surface albedo and
351 CH₄ concentration.

352

353 5 Summary and discussion

354 Remote sensing measurements from airborne and satellite instruments are widely used to detect
355 CH₄ emissions. In our study, the traditional MF and the OE methods are used to quantify the effects of
356 aerosol scattering on CH₄ retrievals based on simulations of AVIRIS-NG measurements. The results
357 show that the retrieval biases increase with increasing AOD and surface albedo for both techniques. In
358 the OE method the biases increase with increasing CH₄ concentration and SSA, but decrease with
359 increasing aerosol asymmetry parameter. The CH₄ retrieval bias increases with increasing XCH₄ in the
360 OE method but decreases for the same scenario in the MF method. The surprising MF trend is attributed
361 to the inability of the MF method to treat nonlinear absorption effects at high XCH₄ values. We also
362 present bias ratios for the two techniques. The MF method shows smaller bias ratios at large CH₄
363 concentrations than the OE method; it is, therefore, the optimal method to detect strong CH₄ emission
364 sources when scattering effects can be ignored in the retrieval. For the same retrieval scenario, the OE
365 method seems to be more suitable for detecting diffuse sources. Further, the MF method relies on a

366 comparison with the background CH₄ concentration. It is difficult to get an accurate estimate of the
367 background XCH₄ value in polluted atmospheric environments. In contrast, the OE method provides
368 retrievals based solely on the atmospheric scenario of interest; CH₄, aerosols and surface albedo can be
369 simultaneously inferred. Therefore, when scattering effects need to be considered, the OE method is the
370 appropriate choice. Indeed, the MF method was intended for plume detection. OE enables accurate
371 quantification of XCH₄ in the presence of aerosol scattering.

372 This study focused on a comparison of retrieval techniques. It is also important to accurately
373 represent the physics of atmospheric RT, especially for scenarios with significant aerosol scattering. RT
374 models traditionally used in retrievals of imaging spectroscopic data use simplified radiation schemes
375 and predefined aerosol models, which may introduce inaccuracies in the representation of atmospheric
376 physics. The 2S-ESS model provides the capability to quantify aerosol impacts on CH₄ retrieval for
377 different aerosol types, optical depths and layer heights. In future work, we will compare retrievals using
378 the 2S-ESS model against those from other commonly used models such as MODTRAN. We will also
379 evaluate the impact of varying instrument spectral resolution and signal to noise ratio for simultaneous
380 retrieval of CH₄, surface albedo and AOD. This will be relevant for the design of imaging spectrometers
381 for planned future missions such as the NASA Surface Biology and Geology (SBG) mission.

382

383 Acknowledgements

384 A portion of this research was carried out at the Jet Propulsion Laboratory, California Institute of
385 Technology, under a contract with the National Aeronautics and Space Administration
386 (80NM0018D0004). V.N. acknowledges support from the NASA “Utilization of Airborne
387 Visible/Infrared Imaging Spectrometer Next Generation Data from an Airborne Campaign in India”
388 program (solicitation NNH16ZDA001N-AVRSNG), and the Jet Propulsion Laboratory Research and
389 Technology Development program. P.K. was funded by the Japan Society for the Promotion of Science
390 International Research Fellow Program. The authors gratefully acknowledge the insightful and
391 constructive comments from the two anonymous reviewers, which improved the clarity and quality of
392 the manuscript, and elevated the significance of the work beyond the original expectation.

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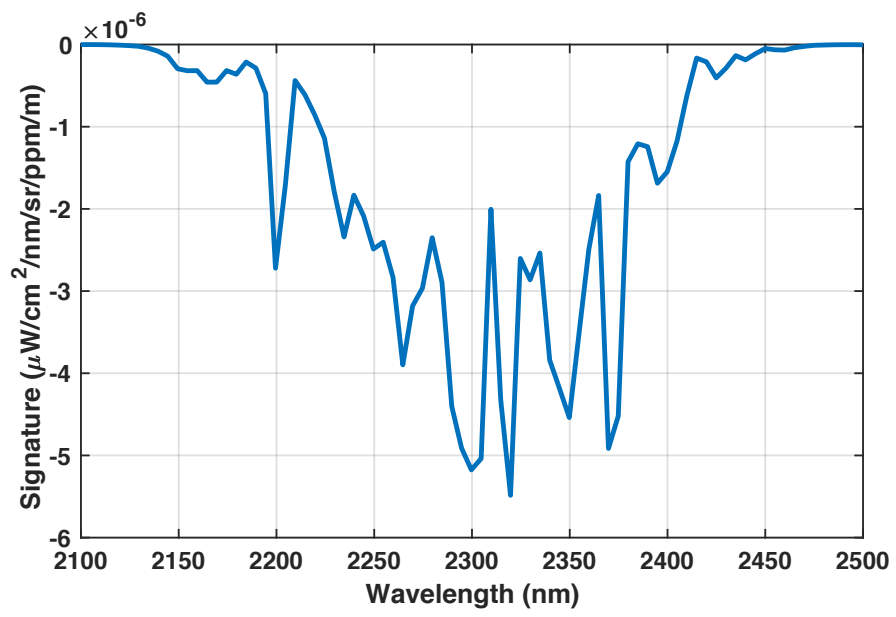
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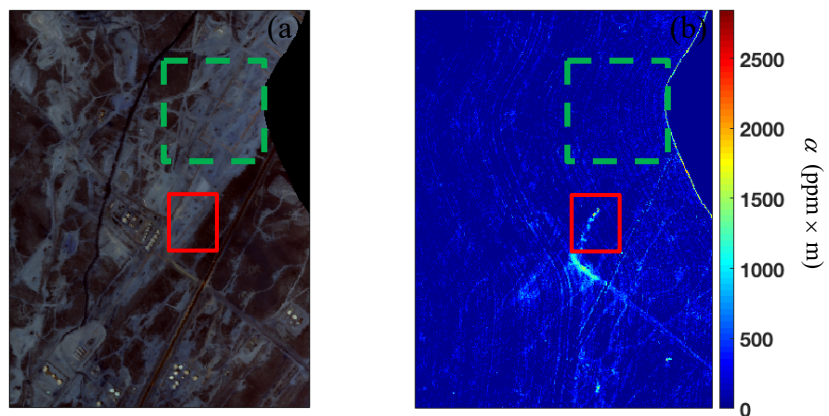
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704 **Figure 1: The target signature used for the Matched Filter method.**

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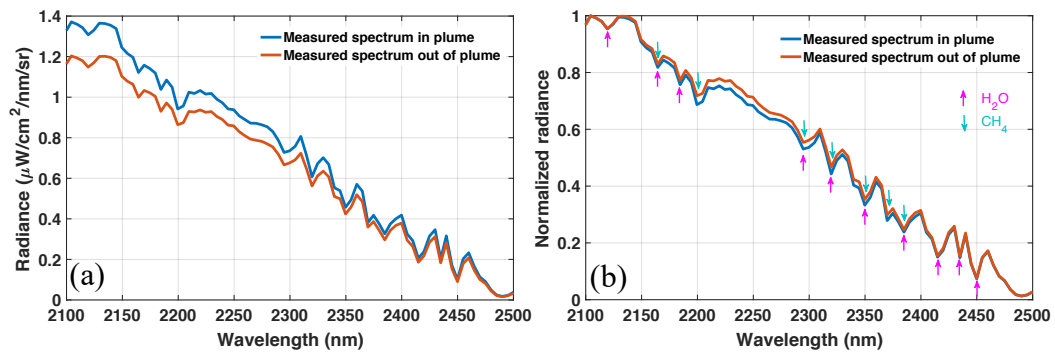


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708 **Figure 2: (a) RGB image of flight data from 4 September 2014 (ang20140904t204546). Adapted from**
709 **Thompson et al. (2015). (b) CH₄ enhancement value α (ppm \times m) obtained by the MF method. An emission**
710 **source is shown in the solid red box and the background region near the target for the MF calculation is**
711 **indicated by the dashed green box.**

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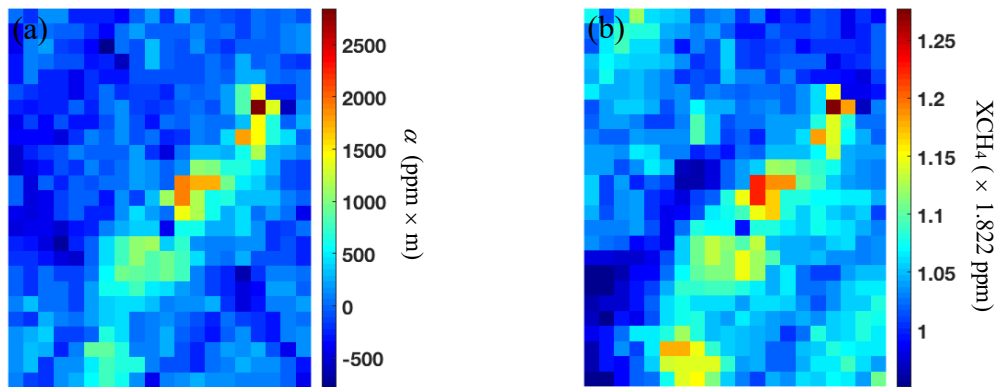
715 **Figure 3: (a) Real radiance and (b) normalized radiance at cross-track detector elements (in and out of plume)**

716 **from the sample AVIRIS-NG measurement. The colored arrows in (b) show the main absorption features due**

717 **to H_2O (purple) and CH_4 (green).**

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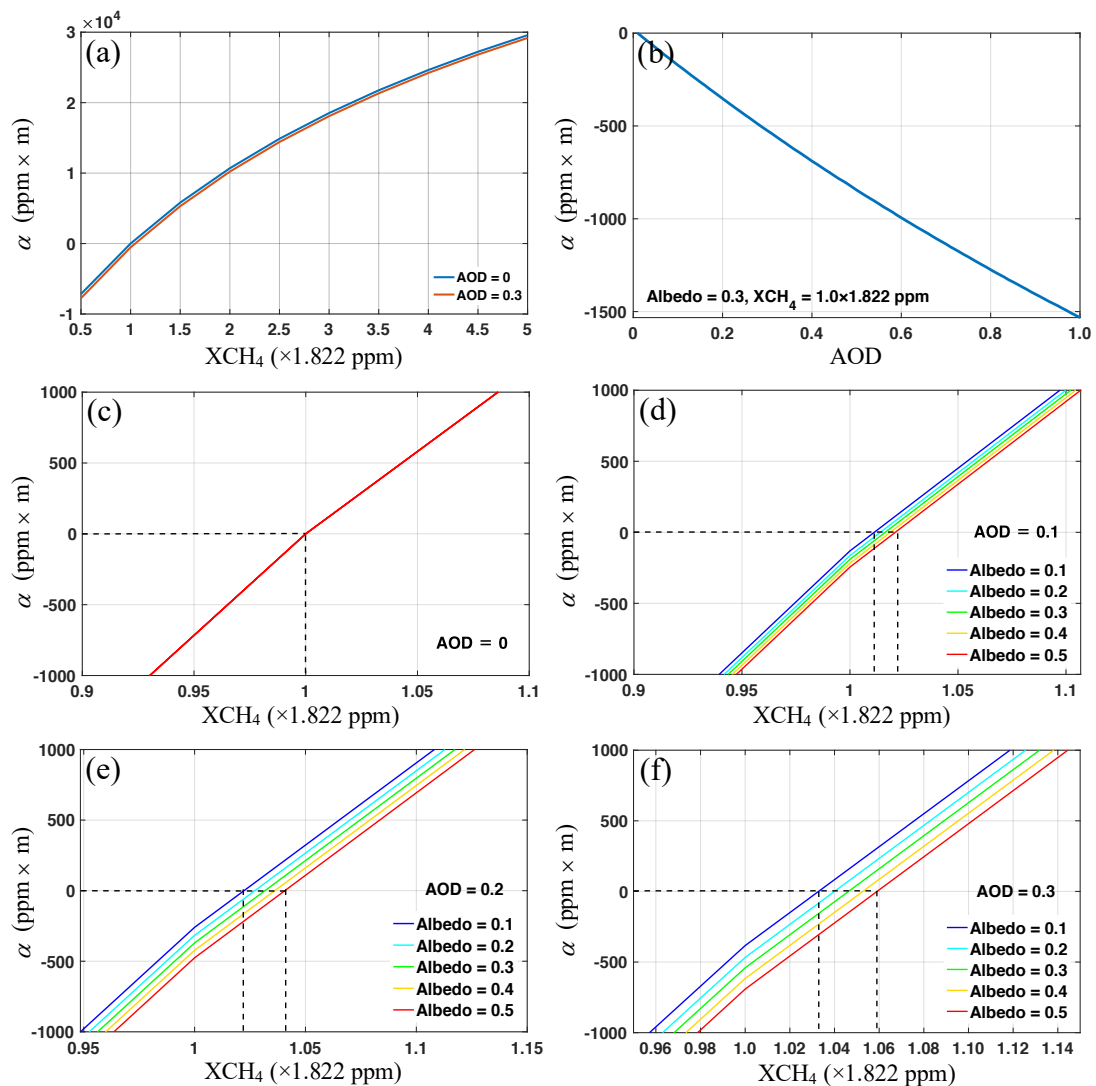
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Figure 4: Retrieval image for the plume center (500 elements) based on the (a) MF method and (b) OE method.

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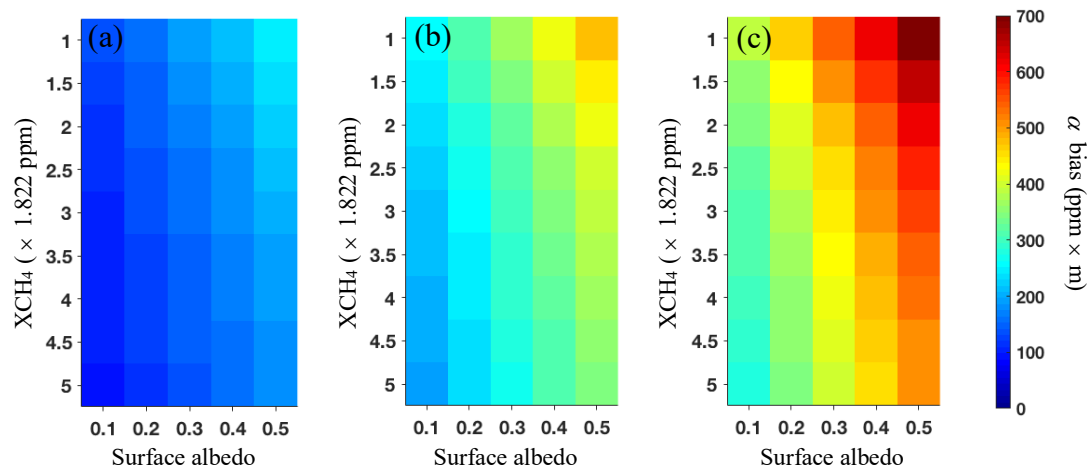
725 Figure 5: (a) α as a function of XCH_4 for AOD = 0 and AOD = 0.3 (surface albedo = 0.3). (b) α as a function

726 of AOD ($XCH_4 = 1.0 \times 1.822$ ppm, surface albedo = 0.3). Zoomed in versions of α as a function of XCH_4 for

727 different surface albedos (0.1-0.5), where (c) AOD = 0, (d) AOD = 0.1, (e) AOD = 0.2, and (f) AOD = 0.3.

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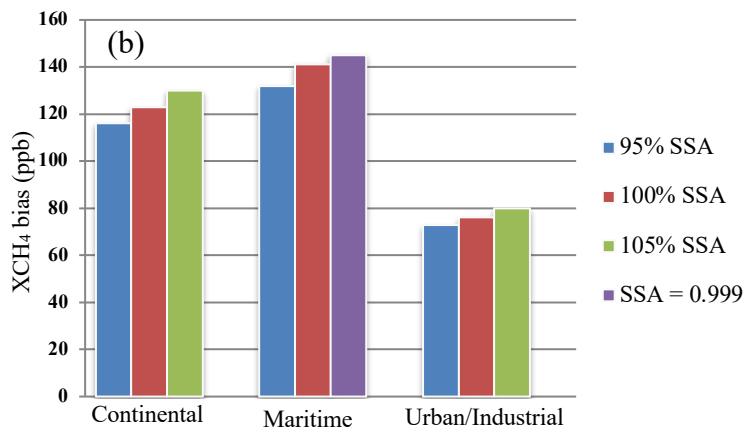
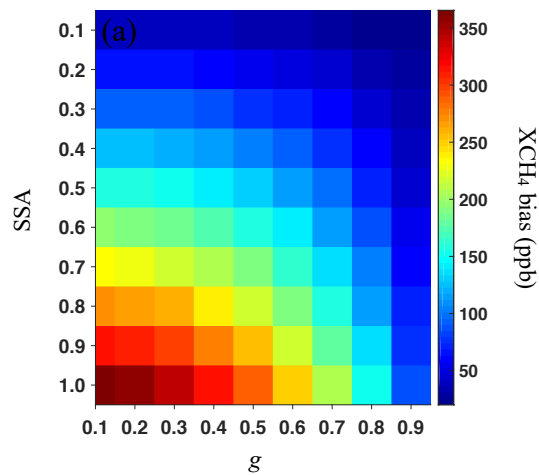
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731 **Figure 6: Bias in α as a function of XCH_4 and surface albedo for (a) $\text{AOD} = 0.1$, (b) $\text{AOD} = 0.2$, and (c) AOD**
732 **= 0.3.**

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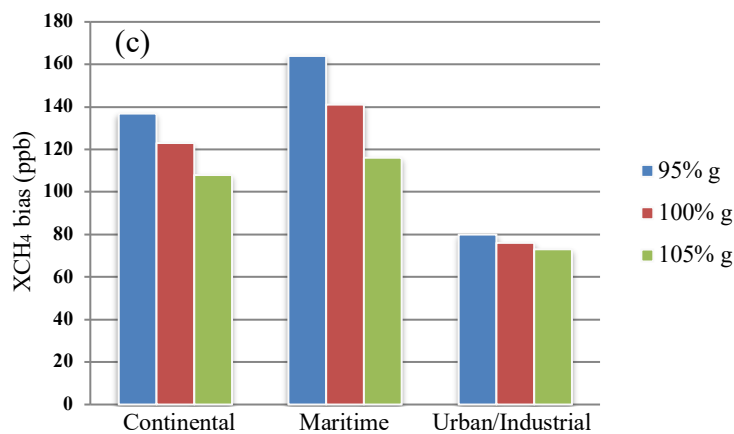
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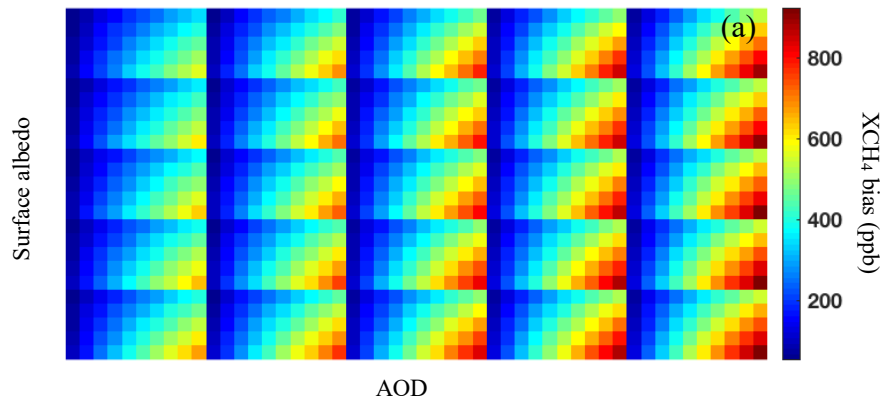
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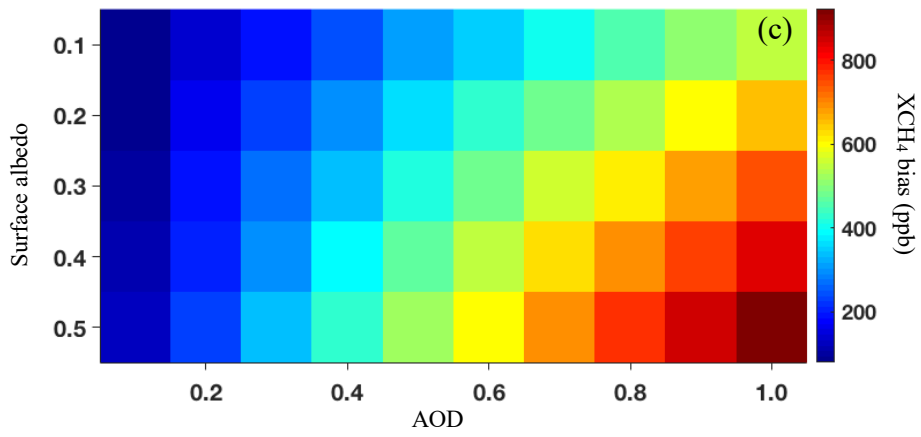
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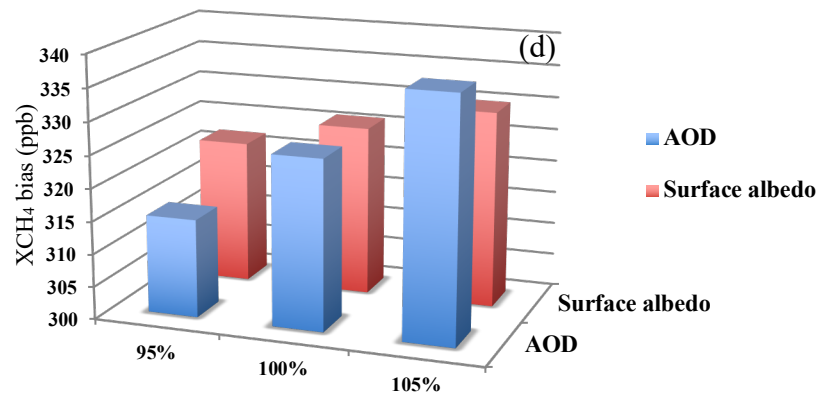
Figure 7: (a) CH₄ retrieval biases for different values of g and SSA. Surface albedo, AOD = 0.3, XCH₄ = 1.0 × 1.822 ppm. (b) CH₄ retrieval biases for a ± 5% change in SSA for the three aerosol mixture models. (c) Same as (b), but for a ± 5% change in g.



1	2	3	4	5 (b)
1.2	2.2	3.2	4.2	5.2
1.4	2.4	3.4	4.4	5.4
1.6	2.6	3.6	4.6	5.6
1.8	2.8	3.8	4.8	5.8

Figure (b) is a table showing the relationship between AOD (x-axis) and XCH₄ bias (y-axis). The color scale ranges from 1 (light blue) to 5 (dark blue) XCH₄ ($\times 1.822$ ppm). The table shows a linear relationship between AOD and XCH₄ bias.





744

745 Figure 8: (a) CH₄ retrieval biases for different values of XCH₄, AOD and surface albedo. $g = 0.75$, $SSA = 0.95$.

746 (b) XCH₄ for each box in (a). (c) Zoomed in plot of bottom right box (XCH₄ = 5.8×1.822 ppm). The x and y

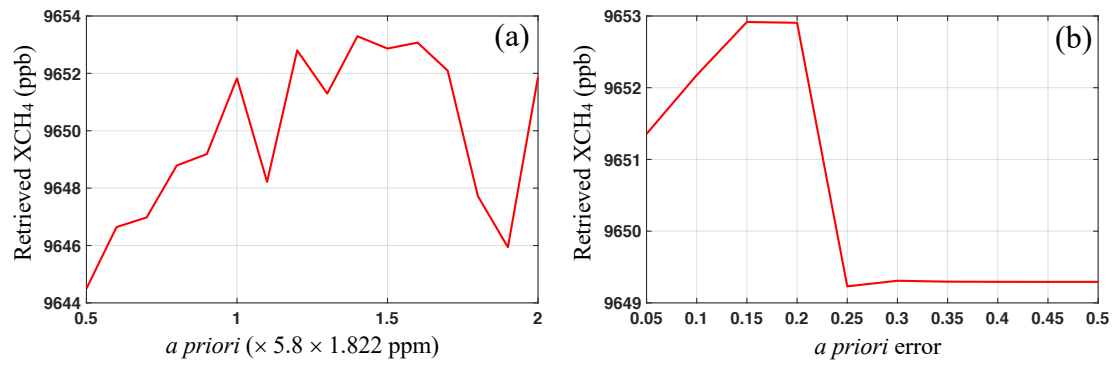
747 axes show the variation of AOD and surface albedo, respectively. These changes are identical for every box

748 in (a). (d) CH₄ retrieval biases for a $\pm 5\%$ change in AOD and surface albedo from a base value of 0.3 ($g =$

749 0.75 , $SSA = 0.95$, XCH₄ = 5.8×1.822 ppm).

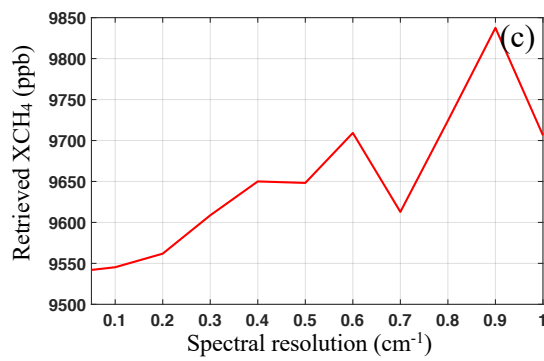
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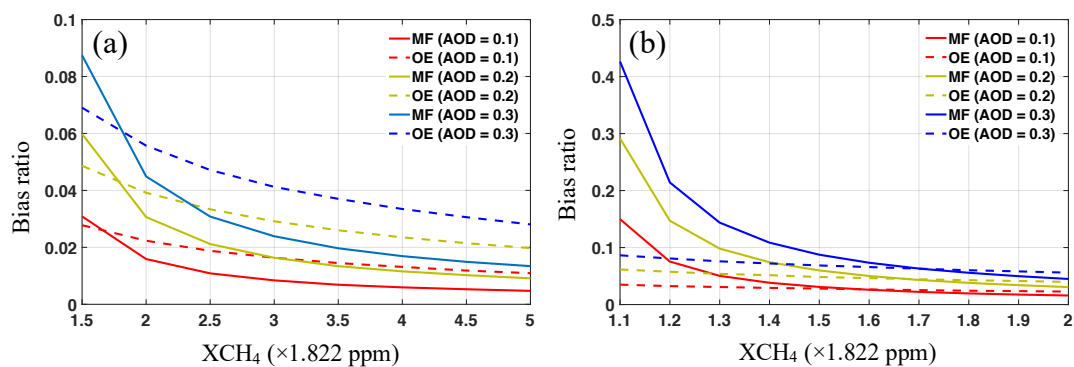


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755 **Figure 9: Retrieved XCH₄ for different values of (a) a priori (a priori error = 0.2), (b) a priori error (a priori =**
756 **5.5 × 1.822 ppm) and (c) spectral resolution. $g = 0.75$, SSA = 0.95, AOD = 1.0, surface albedo = 0.5, XCH₄ =**
757 **5.8 × 1.822 ppm.**

758

759



760 **Figure 10:** (a) Bias ratio as a function of CH₄ concentration for the two retrieval techniques, where the XCH₄
761 ranges from 1.5 to 5 ($\times 1.822$ ppm). (b) Same as (a), but for XCH₄ ranging from 1.1 to 2 ($\times 1.822$ ppm). Surface
762 albedo is set to 0.3 for all cases; results for the MF and OE methods are shown by solid and dashed lines,
763 respectively.

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765

	Dust-like	Water soluble	Oceanic	Soot
SSA	0.805	0.799	0.970	0.014
<i>g</i>	0.926	0.550	0.816	0.092

766

Table 1: Optical properties of basic aerosol types (WCRP, 1986).

767

		Continental	Maritime	Urban/Industrial
Aerosol component	Dust-like	70%		17%
	Water soluble	29%	5%	61%
	Oceanic		95%	
	Soot	1%		22%
SSA		0.746	0.966	0.314
<i>g</i>		0.764	0.810	0.586

768

Table 2: Optical properties of three aerosol mixture models (WCRP, 1986).

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Attribute	Values
Sensor height	1 km
View zenith angle	11.91°
Solar zenith angle	30.75°
Relative azimuth angle	22.87°
Aerosol loading region	surface to 1 km
SSA	0.95
<i>g</i>	0.75

771 **Table 3:** Inputs for the 2S-ESS model simulation.

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