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Comment

## ***Interactive comment on*** “Characterization of methane retrievals from the IASI space-borne sounder” **by A. Razavi et al.**

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### **Note:**

The answers to the referee’s comments are colored in blue and the text updated or changed in the revised manuscript is colored in green. Regarding the figures numbering, we have chosen to use letters for the figures related to this document and to reference the figure in the revised manuscript as “*paper figures*”.

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## 1 Reply to referee 1:

### 1.1 General remarks

The paper "Characterization of methane retrievals from the IASI space-borne sounder" by Razavi et al presents first global total column retrievals using the methane  $\nu_4$  absorption band. The authors also discuss the opportunity to retrieve methane in the 2760cm<sup>-1</sup> region where solar light substantially contributes to the total signal and hence enhances the sensitivity towards lower atmospheric layers. In general, the paper is well written and concise. Further, it is the first to attempt total column retrievals from IASI which is a challenging task with large scientific potential. In some aspects, however, the authors seem to be overly optimistic and their very low error estimates cannot explain the observed features which are, when compared with ground-based concentrations or SCIAMACHY retrievals, partially unrealistic. This concerns some major comments which definitely need to be addressed before being suitable for publication. Given error estimates should be definitely revised as they are misleading and promise a kind of data quality that I really don't see in the global maps.

We thank the referee for his detailed review, useful comments and for sharing his experience on methane sources, variability and on its remote sensing. His major concern is on too optimistic error estimates which are resulting from the OEM. In the revised version of the paper, this has been tackled in a more detailed and systematic manner. Care has also been made not to oversell the IASI  $CH_4$  data product. Furthermore, in order to improve on the comparison with other satellite sounders, we have replaced, in the revised manuscript, the column average vmr of  $CH_4$ , initially calculated with respect to the air density, by another, which includes a correction factor inferred from  $N_2O$ , simultaneously fitted (see below). This procedure, similar to that used for SCIAMACHY processing (where  $CO_2$  is used for the correction), overall decreases the average column vmr by several percents, greatly improving the comparison. We think the paper

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has strongly improved and gained credit by making these changes and we are grateful to both referees for pointing out the loose issues in these first assessments of  $CH_4$  from IASI.

Our point-by-point response to the referee comments is provided below.

## 1.2 Major comments

### all related to the choice of Sa

#### 1.2.1

In figure 4, the a priori covariance (diagonal elements) is depicted. I guess the 1-sigma levels are indicated but this is not clear to me (please clarify). The stratospheric variability in Sa seems to me far too low especially as a global climatology is applied while concentrations at the 15-20km levels at higher latitudes can even be below 1ppm. As the North-South gradient is largely determined by stratospheric variations, the strict choice of Sa will largely dampen the retrievals (or put changes in other height layers).

We thank the referee for these comments on the stratospheric variability. On *paper figure 4*, the grey area corresponds indeed to the  $1\sigma$  levels, this has been clarified in the figure legend and caption. The a priori profile and variability have been derived from a set of LMDZ model profiles gridded on  $2.5^\circ \times 3.75^\circ$ , using 1 day per season only. All the profiles of these 4 days are represented on figure A. As can be seen, the model does not show concentrations below 1ppm between 15 and 20 km which in consequence are absent from the  $1\sigma$  variability in *paper figure 4*. As these low values were measured by limb-sounding instruments [e.g. De Mazière et al., 2008], we agree with the referee that this may bias the retrieval to some extent. However, considering that we start from the same prior at all latitudes, that IASI is hardly sensitive above 15 km and that the UT/LS variability in Sa is small we would rather suggest that

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the North-South gradient observed here is more representative of free tropospheric variations. We agree however, that a stratospheric gradient exist (it is present in the model, amounting about 2.9% (15.5-20 km) but it is there less than the tropospheric gradient which is around 6.8% (0-10km)) and that a different choice of  $x_a$  and  $S_a$  between both hemispheres could change the picture. Further work would be needed to assess this. Details regarding these specific points have been added in section 2.4 of the manuscript:

“Although the same prior information is used for different locations in order to avoid a dependence upon the a priori on the latitudinal scale, it is noteworthy that North-South gradients are present in the troposphere (6.8% from 0 to 10 km) and in the lower stratosphere (2.9% from 15.5 to 20 km).”

## 1.2.2

It would also be better to provide  $S_a$  in its matrix form (eg surface plot) in order to judge the off-diagonal elements. My main concern is the following: I am surprised by the high sensitivity towards surface layers and wonder whether this might be influenced by strong correlations with higher layers introduced by  $S_a$ . How do the averaging kernels change if the lowest 1-2km are decoupled (in  $S_a$ ) from other layers (which is not an unreasonable assumption as methane can accumulate in the boundary layer)? Similar to this remark, it would be useful to add total column kernels in figure 5 and to extend the y-axis to 0 (why does it stop at 2km, this is not clear to me).

The matrix form of the  $S_a$  has been added to *paper figure 4*, as shown in figure B. The revised paper figure caption has been modified into:

“a priori information derived from an ensemble of LMDZ model profiles for four days, each characterizing one season. The values were multiplied by two to increase variability. (a) The a priori profile (yellow curve) and variability (grey area) represented by the standard deviation ( $1\sigma$ ) derived from the covariance matrix. (b) Surface plot of the

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Sa matrix, in %, illustrating both the variability (diagonal elements) and the degree of correlation between the different levels (off diagonal elements)".

The manuscript has been accordingly corrected:

"As the model usually tends to smooth the spatial variability, we have chosen to multiply by 2 the Sa covariance matrix built from LMDZ. Figure 4 presents on one hand the a priori profile and its associated variability limited on both ends by the standard deviation value, and on the other hand the surface plot of the Sa matrix which also highlights the a priori vertical correlation between the layers. It can be seen that the model Sa matrix show a large correlation length (about 8 km), with off-diagonal elements that still make up half of the ground variability in the middle troposphere."

The low degree of vertical information is furthermore obvious from the averaging kernels (figure C, left panel), which essentially shows that the sensitivity at the surface comes from layers above (e.g. the 0-3 km kernel peaks around 8 km). The vertical correlations in  $CH_4$  are expected to be important globally due to the long lifetime but we agree with the referee that this does not hold locally, on occasions where  $CH_4$  accumulates in the boundary layer above source regions. In order to test to what extent the low vertical resolution relates to the choice of Sa, we have made a comparison of the retrieval results using the LMDZ Sa matrix with a slightly correlated Sa matrix (1.5 km correlation length only). The comparison of the resulting averaging kernels, provided in figure C, tends to show that the vertical sensitivity is inherent to the measurement and not significantly affected by the choice of Sa: indeed there is only minor changes to both the vertical resolution and the surface sensitivity. This has been added in the revised manuscript (in section 2.4: "Preliminary tests suggest that using a different covariance matrix with uncorrelated layers does not significantly increase vertical sensitivity."). We should point out here that the retrievals correspond to daytime measurements where the thermal contrast is not higher than 3 K, not enhancing the surface sensitivity. Finally, the sensitivity is given for the lowest retrieved layer (0-3 km), which is at 1.5 km and thus not exactly representative of surface sensitivity. The latter is even smaller, probably close to zero. The sentence "Note that the averaging kernels are

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plotted against the middle of the retrieval layers in such a way that the first point is located at 1.5 km (0-3 km layer).” has been added to the manuscript.

From the different tests achieved, it seems unlikely to extract different tropospheric partial columns from IASI measurements in  $\nu_4$ , justifying our choice of providing a total column only (though with mid tropospheric maximum sensitivity).

The total column averaging kernel has been added to *paper figure 5* (corresponding to figure D) as proposed by the referee. And the paper figure caption has been updated into “Averaging kernels presented in mixing ratios unit for representative cases of (a) tropical, (b) midlatitude and (c) polar regions. The black curves represent the total column averaging kernels, with absolute values given by the top scale. The averaging kernels rows are plotted with respect to the middle of the retrieval layers.”

### 1.2.3

Figures 7 and 10 show actual retrievals, with values up to 2.1ppm. If I read the  $S_a$  values correctly, this would be up to 3-5 $\sigma$  higher than the prior. Hence, the retrieved state is far away from the prior and the retrieval tries to just shift the profile as this minimises the penalty introduced by  $S_a$ . This can be seen in Fig. 10 where the retrieved profile seems uniformly shifted by about 200ppb (ie more than 2 $\sigma$ ). Either, the retrievals are strongly biased (which seems more realistic) or the prior is chosen too low and too conservative in terms of variations. Ground-based retrievals in Alert, for instance, are on average about 1850ppb and seldom exceed 1900ppb. Retrieved values of up to 2ppm in the total column at high latitudes therefore seem unrealistic and cannot be reconciled with the 1-1.5% error estimate for the total column.

We thank the referee for these comments on the absolute values of the methane vmr (see also discussion of the global distribution below). Although this paper is not validation-oriented, we really ought to have checked this before. The large values in

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the paper came from the fact that the column averaged  $CH_4$  vmr was obtained by normalizing the retrieved column simply by the air column, the latter being provided by the operational level2. In the revised manuscript we have changed the procedure of calculating this quantity, following a methodology similar to that used in SCIAMACHY. Here, we have chosen to normalize the retrieved values with respect to  $N_2O$ , retrieved simultaneously in the spectral range. The following formula was applied:

$$vmr_{CH_4}^{ret.} = \frac{TC_{CH_4}^{ret.}}{TC_{N_2O}^{ret.}} vmr_{N_2O}^{ref.} \quad (1)$$

Where  $vmr_{N_2O}^{ref.}$  is the  $N_2O$  vmr given in the 2007 IPCC report. This procedure is valid given the very long lifetime (114 years) of that species and it permits that the mutual errors cancel each other out. *Paper figure 8* in section 3.2 (corresponding to figure E) and *paper figure 11* in section 3.3 were revised accordingly.

*Paper figure 8* now shows column-averaged mixing ratios varying, except for very large (unrealistic) values found around the Antarctic, between 1.65 in the tropics and 1.88 ppm at northern mid and high latitudes. These values are consistent with literature data. In particular the comparison is in all respect good with AIRS while differences in the strength and location of the North-South gradient with SCIAMACHY remain. This discussion is also expanded in the manuscript (section 3.2). The revised paper figure and the text have been changed according to these new results:

“In order to provide a distribution which does not depend on the ground altitude and where retrieval errors are minimized, the global distribution is expressed in normalized vmr, with the  $CH_4$  total columns divided by the  $N_2O$  total columns, simultaneously retrieved. This procedure is similar to the SCIAMACHY methodology where the methane column was divided by the  $CO_2$  column [Frankenberg et al., 2006]:

$$vmr_{CH_4}^{ret.} = \frac{TC_{CH_4}^{ret.}}{TC_{N_2O}^{ret.}} vmr_{N_2O}^{ref.} \quad (2)$$

Where  $vmr_{CH_4}^{ret.}$  is the methane column averaged vmr,  $TC^{ret.}$  is the retrieved total col-

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umn of methane or nitrous oxide and  $vmr_{N_2O}^{ref} = 319 \text{ ppb}$  is the reference mixing ratio of  $N_2O$  as given in [Forster et al.,2007].

The global distribution (see Fig. 8) is illustrated on a  $4 \times 4^\circ$  grid with a projection chosen to highlight the northern hemisphere. High values found over the Antarctic are rejected by posterior filters but some unrealistic large values above these cold regions remain (not shown on the figure). Excluding these extremes values, the methane normalized mixing ratios range from 1.65 in tropical regions up to 1.88 ppm at high northern latitudes with a distinct North-South gradient. The cut-off from low to high concentrations is located around  $20^\circ\text{N}$  with a further steady increase towards the polar region. The elevated methane concentrations at mid and high latitudes could be related to the locations of large methane emission sources (such as rice agriculture, livestock and wetlands).

Comparing the global distributions of total columns with other satellites is a difficult task, mainly because of the vertical sensitivities inherent to each observing modes. For example, regarding the AIRS distribution [Xiong et al., 2008], the observations are expressed in mixing ratios for two vertical layers (150–250 hPa and 450–550 hPa) averaged for the month of August 2004. Given the IASI vertical sensitivity, the comparison with the layer between 450 and 550 hPa is more relevant. The range of IASI methane column averaged vmr are in good agreement with the AIRS mixing ratios corresponding to partial columns (extending from 1.65 to 1.9 ppm) and the North-South gradient is similar in strength and location. The tropical IASI distributions shown in [Crevoisier et al., 2009] give as well similar concentrations. However, our methane averaged column present lower values in the tropics (about 20 ppb) than both products, indicating the possible contamination by water vapor in humid regions. Concerning SCIAMACHY, the methane product is an averaged vmr more representative to the lower troposphere than the IASI one [Frankenberg et al., 2008b]. The concentration range is therefore slightly different (1.63 to 1.81 ppm for SCIAMACHY) and only enables the qualitative comparison between the two global distributions. Doing this, we observe that the lat-

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itudinal gradient from the yearly averaged distribution of SCIAMACHY is sharper and located farther south. It also points out strong methane emissions in various part of the world (such as Southeast Asia and central Africa) which are not detected by IASI, probably due to its lack of sensitivity close to the surface.”

In *paper figure 11*, the retrieved profiles correspond now to a sunglint contaminated spectrum (see a few comments below) and has similarly been normalized with respect to  $N_2O$ , considering its reference vmr. This Figure now shows profiles which remain within the  $2\sigma$  variability of Sa around the a priori profile.

#### 1.2.4

Considering the strict Sa and deviations from the prior: It would be worthwhile to investigate how a relaxation of Sa (or shift in  $x_a$ ) change the fit residuals. Given the large deviations from the prior, the penalty induced by this deviation might influence the fit residuals as the retrieval becomes too constrained. Furthermore, the posteriori error estimate is (in case the retrieved state is far away from the prior) largely constrained by Sa.

As suggested, we have tested the influence of a shift of the methane a priori profile (+100 ppb) on a few orbits of IASI nadir measurements. The difference between the rms is not significant at high-latitudes, but somewhat better at middle latitudes and in the tropics. Regarding the retrieved vmr, there are now larger by 40 ppb on average but always lower than the prior, suggesting a poor representation of the latter. As will be discussed below, it is difficult to place an exact value for the error on the retrieved column but this simple test, suggested by the referee, already points to the fact that similar fits quality can be achieved with methane column differing by 2%. Although the retrievals are less likely for the test conducted here (because  $x_a$  is out of the range of the retrieved columns), we agree that our 1% error is overstated when only accounting for profiles within the statistics. This has been corrected throughout the manuscript

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(section 3.1: “The retrieval error of the methane total column (from 0 to 21 km) resulting from the OEM can be as small as 1%. However, the value of the column error is intimately related to the a priori variability and correlations between the levels of the profile. Considering that much larger variability can occur in the boundary layer nearby source regions, where IASI is less sensitive, this value is obviously a lower bound to the error; in fact it is only valid for cases which are included in the ensemble of profiles used to built the a priori matrix. Other cases, and in particular those showing enhancement in the boundary layer will not be reproducible with this accuracy.” and section 4: “The retrieval error (which only accounts for the smoothing error, measurement error and the error from the humidity profile) estimated in this study is to be considered as a lower bound to the error in case of background methane profiles. As discussed in Sect. 3.1 a more quantitative evaluation of the accuracy associated with the IASI  $CH_4$  product will only be possible by performing a large validation exercise, which is outside the scope of this paper.”)

### 1.2.5 sensitivity to the total column

There is another IASI paper on ACPD by Crevoisier et al. One sentence reads: "Hence, IASI only allows the retrieval of a mid-to-upper tropospheric integrated content of methane". Please discuss why your retrievals are sensitive to the lowest layers (ie what is conceptually different in the approach) and how you manage to obtain an error as low as 1% for the total column. In particular, I would like to see evidence that boundary layer enhancements can be detected by IASI. For this purpose, you could even perform a synthetic retrieval with enhanced methane only in the 0-2km layer (say 200ppb higher than the prior). Would IASI really detect this signal or is the high sensitivity to this layer artificially introduced by Sa?

The methane product presented in our paper is derived from the retrieval of a total column (i.e. a shift of the a priori profile) but the sensitivity of this retrieval is principally

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resulting from the mid-troposphere (4-12 km) as seen in the averaging kernels on *paper figure 5* (or figure D). Our vertical sensitivity is thus fully consistent to the one described in Crevoisier et al, furthermore pointing to the relatively minor influence of Sa. This has been clarified in the abstract: “In this paper, we present the first global distribution of methane total columns (mostly sensitive to the middle troposphere) from the IASI spectra using the methane  $\nu_4$  absorption band” and throughout the manuscript (in section 3.1: “It follows that we consider only the total columns for the derivation of global distributions. The column is mostly driven by the free troposphere where IASI sensitivity is highest.” and in section 3.2: “In this section, we show the preliminary global distribution of methane total columns assessed from the IASI sounder; as explained before the columns are representative of the middle troposphere (see the averaging kernels on Fig. 5)”.

In order to get a better estimate of the error (the 1% provided is a retrieval error only, for cases that fall within the statistic introduced by Sa) synthetic spectra have been simulated with a 200 ppb enhancement of methane in the boundary layer, as suggested (we thank the referee for this). Figure F represents the results of the forward model simulations, plotted as the difference between the spectrum with enhanced  $CH_4$  (200ppb) and the spectrum corresponding to the a priori. The differences are shown for several values of the thermal contrast (the difference between the ground temperature and the temperature of the first atmospheric layer). Comparing these to the IASI noise shows that the instrument will be blind to the 10% enhancements as long as the thermal contrast remains below 5K. Above this value, the difference emerges from the noise and should be detectable, although the concentration will still be underestimated. Thermal contrast in the range +10 to +20 K are frequently valid for IASI overpasses in the morning but not in the evening, where they are close to zero (or even negative).

This figure has been added to the manuscript (*paper figure 7*) together with the following description: “In order to better show this, we have tested the IASI capability to detect a  $CH_4$  source region with forward model simulations. Figure 7 provides the transmittance differences in the spectral range selected between the methane a priori

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profile and a 200 ppb methane increase in the boundary layer. Simulations with different values of thermal contrast (i.e. the difference between the ground temperature and the temperature of the first atmospheric layer) from +0.5 to +20 K have been performed. The comparison between these simulations and the IASI noise (green curve) shows that IASI should detect the 10% increase of  $CH_4$  for cases when the thermal contrast exceed 5 K although the constrain of the Sa matrix will induce some under-estimation. A full error characterization will have to be evaluated at a later stage by a detailed validation exercise.”

It is very difficult at this stage to get a very precise error estimate of IASI  $CH_4$  retrievals above source regions, as this will be strongly related to both the strength of the enhancement and the local thermal contrast situation. We fully agree with the referee, however, that the 1% value reported is far too optimistic for non-background situations and can be misleading in the interpretation of the data. In the revised manuscript, we have changed the 1% value whenever necessary (see reply 1.2.4).

### 1.2.6

The potential of IASI is promising but is is rather bold to claim 1% error in the total column while there are unrealistic values as high as 2.1 ppm. Results should be compared with optimised model fields (eg optimised to ground-based stations) in order to draw any preliminary conclusion on errors. As far as I can judge, the choice of Sa and some unaccounted systematic errors lead to an unrealistically low a posteriori error estimate.

We agree with all these comments, which we think have now been adequately answered in the previous comments.

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## 1.2.7 Figure 7

Please change to another projection that allows to see the entire earth. Why are the tropics so low even though SCIAMACHY, IASI (in Crevoisier et al) and atmospheric models (mainly because the tropics are well mixed with high values throughout the column up to the high tropopause)? Is there a problem with water vapor interferences (as discussed and eliminated in Crevoisier et al)? Surprisingly, the colorscale ranges from 1840-2000ppb which is inconsistent with Crevoisier et al results, SCIAMACHY retrievals, ground-based measurements and TM5 model fields (eg Schneising et al, ACP, 2009). Basically, your lowest values are even higher than the highest values in other retrievals. How can this be explained?

See above for the discussion of *paper figure 8* and the change of procedure to calculate the column average mixing ratio, which now reconcile the IASI retrievals between them (see Crevoisier paper in this issue) and also with other satellite and model data. Even proceeding this way, we find very large and unrealistic values circling the Antarctic, which point to radiative transfer issue. Furthermore, the low radiance signals above the Antarctic continents resulted in large errors which have been filtered out. For these reasons, and unless the referee objects, we prefer keeping the polar projection in the paper. However, we now make explicit mention of the high values around the Antarctic.

## 1.2.8 Page 7626, lines 10+

Again, I consider it rather bold that the features with high values correlate with regions of typical methane emissions. I don't think this statement is justified (what happens over the oceans, why are the values way too high, why don't you see stratospheric depletions in polar air??)

The values above the ocean are now lower with the  $N_2O$  normalization. In fact the highest values are now located above Siberia, which is interesting and could stimulate

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further detailed and focused analyses there.

### 1.3 Specific comments

#### 1.3.1 Page 7616, line 24

1774.62+-1.22: I would skip the last two digits.  
[corrected](#)

#### 1.3.2 Page 7616, line 17

large uncertainties still exist.  
[corrected](#)

#### 1.3.3 Page 7628, lines 8+

is an increase of 0.06 DOF really significant? The potential of adding this spectral region is enticing but from Fig 10 it looks as if both retrieval windos are not yet consistent. [The point we want to make is more on the level of vertical sensitivity enhancement near the surface. However, another case has been chosen where the increase of DOFS is higher \(0.24\). This new observation corresponds to a sunglint contaminated spectrum recorder above the Indian ocean. \*Paper figures 10 and 11\*, as well as the text in section 3.3 have been accordingly changed. The results presented in this section are preliminary and more work above different surface types and with different viewing angles \(e.g. different reflectances\), etc is obviously needed to fully gauge the potential of using this additional band. The following sentences have been added to section 3.3:](#)

["In order to fully determine what the methane retrievals in the  \$\nu\_3\$  band have to offer, more tests must be conducted. We expect to observe a larger signal for off-nadir](#)

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recorded spectra as the angle between the sun and the satellite plays a significant role in the reflectance value. However, due to the decrease of the surface temperature, lower sensitivity is expected at high latitudes.”

#### 1.3.4 Figure 10

which location is this retrieval? You could compare surface values with NOAA GMD measurements (via the interactive data viewer). I am pretty sure you will not find 2.0ppm.

As explained above, a new example has been chosen to illustrate the sensitivity enhancement capability of IASI with the  $\nu_3$  methane band. The profiles presented before were retrieved from a spectrum recorded above a desert area where spectrally dependent emissivities introduced some errors.

The nearest NOAA site is now located in Seychelles, Mahe Island ( $4.67^\circ S$ ,  $55.17^\circ E$ ) and provide a surface value of around 1810 ppb in January 2009. The surface concentrations found on *paper figure 11* (corresponding to figure G) are smaller. This could be explained by the interference of water vapor in this very humid region. This consideration has been added to the manuscript:

“Care should be taken in interpreting these values as high water vapor concentration are present in this region, which could bias the methane retrieval (as mentioned in the global distribution description), figure 11 is in itself illustrative of the relative sensitivities of the  $\nu_3$  and  $\nu_4$  bands to the vertical structure.”

## 2 Reply to referee 2:

In general, the paper “Characterization of methane retrievals from the IASI spaceborne sounder” by A.Razavi et al. is well written to characterize the methane retrieval from

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IASI, and discuss the opportunity to use shortwave band near 2760 micron to increase the sensitivity to the surface methane. However, the authors oversell the product by claiming the error of 1% in the total column. Since the maximum sensitivity is between 4-10 km and the sensitivity below 2 km is low, and the profile error is between 1-2.5%, from my it is impossible to derive the total column in 1%. The VMR of 2.101 pm from global distribution and mixing ratio about 2 ppm in Figure 10 is way too high than reality, indicating some problems in the retrieval. Moreover, this is inconsistent with the claimed error of 1%. This concern has to be addressed and clarified before being suitable for publication in ACP.

We acknowledge the referee for his/her very useful comments which have greatly helped to improve the results presented in the manuscript. The latter has been strongly revised in several respects to deal with the above-mentioned issues, also raised by the other referee. The two major points (error estimate and overestimation of the total columns) are discussed hereafter:

## 2.1 Error estimate

Regarding the 1% error on the total column, this is the retrieval error (smoothing and measurement error) resulting from the use of the optimal estimation method. We fully agree with the referee that this error is certainly underestimated, at least for cases that fall outside the statistics included in Sa. Among these cases are all the situations where boundary layer enhancement will be higher than representative global variability. In order to check this we have added, as suggested by the other referee a new figure (figure E corresponding to *paper figure 7* in section 3.1), which highlights the capability of IASI to identify a 10% increase in the boundary layer, under various thermal contrast situation. Figure E represents the results of these new forward model simulations, plotted as the difference between the spectrum with enhanced  $CH_4$  (200 ppb) and the spectrum corresponding to the a priori. Comparing these to the IASI noise shows that

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the instrument will be blind to the 10% enhancements as long as the thermal contrast remains below 5K. Above this value, the difference emerges from the noise and should be detectable, although the concentration will still be underestimated. Thermal contrast in the range +10 to +20 K are frequently valid for IASI overpasses in the morning (over land) but not in the evening, where they are close to zero (or even negative). The description of *paper figure 7* added to the manuscript can be found in reply 1.2.5.

Although it is very difficult at this stage to get a very precise error estimate of IASI  $CH_4$  retrievals above source regions, as this will be strongly related to both the strength of the enhancement and the local thermal contrast situation these simulation indeed point to the fact that the 1% value reported is far too optimistic for non-background situations and can be misleading in the interpretation of the data. In the revised manuscript, we have changed the 1% value whenever necessary (see end of reply 1.2.4).

## 2.2 Absolute value of the $CH_4$ column in the global distribution

The global distribution given in the initial manuscript was biased to too high absolute values in the column averaged mixing ratio. This comes from the fact that this quantity was calculated by normalizing the retrieved column by that of air, not accounting for possible problems in the radiative transfer, which, for methane, rapidly become crucial. In the revised manuscript, we have changed the procedure of calculating the column averaged mixing ratio, following a methodology similar to that used in SCIAMACHY. Here, instead of correcting the column by  $CO_2$ , we have chosen to normalize the retrieved values with respect to  $N_2O$ , retrieved simultaneously in the spectral range. The following formula was applied:

$$vmr_{CH_4}^{ret.} = \frac{TC_{CH_4}^{ret.}}{TC_{N_2O}^{ret.}} vmr_{N_2O}^{ref.} \quad (3)$$

Where  $vmr_{N_2O}^{ref.}$  is the  $N_2O$  vmr given in the 2007 IPCC report. This procedure is valid

given the very long lifetime (114 years) of that species and it permits that the mutual errors cancel each other out. *Paper figure 8* in section 3.2 (corresponding to figure E) and *paper figure 11* in section 3.3 were accordingly revised.

*Paper figure 8* now shows column-averaged mixing ratios varying, except for very large (unrealistic) values found around the Antarctic, between 1.65 in the tropics and 1.88 ppm at northern mid and high latitudes. These values are much more consistent with literature data. In particular the comparison is in all respect good with AIRS while differences in the strength and location of the North-South gradient with SCIAMACHY remain. This discussion is also expanded in the manuscript (section 3.2). The revised paper figure and the text have been changed in section 3.3 according to these new results: see reply [1.2.3](#).

In *paper figure 11*, the retrieved profiles correspond now to a sunglint contaminated spectrum (see a few comments below) and has similarly been normalized with respect to  $N_2O$ , considering its reference vmr. This figure now shows profiles which remain within the  $2\sigma$  variability of Sa around the a priori profile.

### 2.3 Major comments

It is not clear the use of a priori information in different regions, particularly the off-diagonal elements in the covariance matrix, as it is related closely related with the adjust of the lower troposphere  $CH_4$  as seen in Figure 10. Otherwise, the error of 1% is impossible to reach.

The same a priori information is used globally in order to avoid a dependence upon the a priori in the analyses. Given the limited vertical resolution achieved, of course off-diagonal elements play a big role in retrieving profile information. A surface plot of the covariance matrix has been added to figure D in order to explicit the off-diagonal elements. The model Sa matrix show a large correlation length (about 8km) with off-

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diagonal elements that still make up half of the ground variability in the upper troposphere. The new description has been added to section 2.4 for clarification (see reply 1.2.2).

## 2.4 Specific comments

### 2.4.1

Given the sensitivity in the Q branch is much larger than in the spectral window selected for retrieval, it is hard to believe to the impact to include and exclude Q branch channels on the retrieved profile was insignificant. More tests are recommended.

It is true that the exclusion of the methane Q branch decreases the vertical sensitivity and that it can accordingly impact on profile retrievals. However, as there remain some spectroscopic issues for that particular feature, and as we show the column result not to be significantly affected by taking the Q branch into account or not, we have chosen to keep the more conservative approach that excludes this critical range to the detriment of vertical resolution.

### 2.4.2 Page 7624, lines 5-6:

It is not clear “the same priori information is used for each location and time”. Does the priori information for different location change? For instance, are the priori file as well as its covariance are the same in the northern and southern hemisphere?

Yes, there is a single a priori profile and covariance matrix used globally (see reply of the major comment).

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2.5 Page 7624, lines 16:

the maximum sensitivity in the tropics is between 8-14 km, instead of 4 to 10 km.

The sentence has been updated into “It follows from the shape of the averaging kernels that the IASI measurements have a maximum sensitivity to methane in the middle troposphere, between approximately 4 and 10 km for mid and high latitudes and between 7 and 14 km for tropical regions.”

2.5.1 Page 7624, lines 20-21:

this sentence is not complete.

The sentence has been changed into “The resulting DFS for the 3 scenes shown in Fig. 5 are equal to 1.16, 1.04 and 0.92, respectively (for example, this is smaller than CO where the DFS frequently reaches values above 1.5, see [Turquety et al., 2009]).”

2.5.2 Page 7625, lines 27-28:

what does it really mean “error” exceeding 1.5% ? what is the truth it is compared ? need to revise it.

The error on the total column is the relative error expressed in percent (i.e. the retrieval error divided by the methane total column x 100). As explained above this error represents the retrieval error (measurement and smoothing), as calculated in the Optimal estimation method. It is thus only representative for cases which fall inside the global statistics built from models. We fully agree that these error estimates are over-optimistic and that much larger differences will undoubtedly be found when comparing to true correlative measurements. We have tackled this issue in part by adding a series of forward model simulations that characterize the sensitivity to a boundary layer enhancement, which is the larger bias source considering the rather weak IASI sensitivity

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to the surface. Furthermore, we have been more careful throughout the manuscript in not overselling the product by stating the error is around 1% (see reply regarding the error estimate).

### 2.5.3 Page 7626, line 10:

normalized mixing ratio of 2.101 is unrealistic. Some thing must be wrong here See reply 2.2. We think that this issue was handled successfully by changing the normalization procedure, using the  $N_2O$  column retrieved simultaneously as the  $CH_4$  column, instead of the air density. Proceeding this way reduces radiative transfer/retrieval errors, and greatly improves the comparison with other satellite distributions.

### 2.5.4 Page 7626, line 11:

the cut-off latitudinal gradient at 30 N. Is it realistic ?

Page 7626, lines 15-19: looks the error is associated with the altitude, not just emissivity.

Page 7626, lines 26-27: “. . . SCIAMACHY is sharper and located farther south”. Is it the slope of gradient from SCIAMACHY is larger ? why located farther south ? Not clear.

We see in figure E an obvious difference between lower values at the equator and higher values polewards. There is a clear cutoff in the Northern hemisphere between the tropical regions and the mid to high latitudes. We do not see important problems with altitude, in the revised figure (improved normalization with  $N_2O$ ), but emissivity issues remains important above desert area. These emissivity scenes were mostly filtered out, leaving white areas without measurements on the maps (e.g. Sahara).

Regarding the comparison with SCIAMACHY we still find slightly higher concentrations and a different North-South gradient location. This and the comparison also with AIRS

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has been added in the manuscript (see reply 1.2.3).

2.5.5 Page 7628, lines 8-10:

is an DOF increase of 0.06 significant ? The difference in Left and right (Fig 9) is very small. How about in the mid latitude or high latitude region ?

The 0.06 on the DOFS is low but still could be useful for source identification. We have however chosen a new example which shows a larger increase of DOFS (0.24) considering the retrieval in both bands as compared to the retrieval in the  $\nu_4$  band alone. We think that this new example using a sunglint contaminated spectrum will illustrate more strongly the potential of the  $\nu_3$  band. The section 3.3 and *paper figures 10 and 11* have been changed accordingly.

We agree with the referee that more work above different surface types and with different viewing angles (e.g. different reflectances), etc is obviously needed to fully gauge the potential of using this additional band. We have therefore added a paragraph to section 3.3 (see reply 1.3.3).

2.5.6 Fig 10:

Since the sensitivity using  $\nu_3$  is low in the lower troposphere (below 2 km), why does it move even larger than its most sensitivity region at 10-14 km? Some changes must have been made.

As our example was a bit unfortunately chosen because of emissivity features, we have chose to analyze another spectrum with higher reflectance.

Although the effect described by the referee is explainable by looking at the Sa covariance matrix (see figure B), which shows larger variability at the surface as compared to the free troposphere, the updated figure corresponding to our new case (figure G or *paper figure 11*) shows that the retrieval of the  $\nu_3$  band is at larger distance of the a

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priori profile than the retrieval using the  $\nu_4$  band.

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