Response to Reviewer #2

The authors would like to thank the reviewer for his/her interesting and helpful comments. Our answers to all of them are listed below. The comments of the referee are marked in black the answers are marked in blue.

This is a review of the paper titled, "Retrieval of near-surface sulfur dioxide (SO2) concentrations at a global scale using IASI satellite observations", by Bauduin et al. 2015. In general the paper is well written with a good description of the IASI SO2 retrieval methodology. One main comment that affects many parts of the analysis and interpretation of the new results is the lack of information on the detection limit and sensitivity of the IASI SO2 observations. This needs to be demonstrated and made more transparent in the paper.
The sensitivity of the IASI SO_2 observations are discussed in detail in the manuscript, please see in particular Figure 4 and the related discussion.

Comments:

1) One main comment that would greatly benefit the SO2 research community in terms of using these SO2 IR observations would be to provide more insights on the sensitivity and information being provided by IASI on SO2. Some related questions and comments around this issue include:

    a. What do these new IASI global SO2 satellite near surface (anthropogenic) observations provide that are not already being provided by UV/VIS (i.e. OMI, and it the future TropOMI and Tempo)? For example, if IASI is not as sensitive as UV/VIS satellite observations under most atmospheric conditions, however, under ideal IR viewing atmospheric conditions this new retrieval approach provides similar information this should be clearly stated in the paper. Also, is there anything that the IR can (or potentially can) provide that the UV/VIS cannot in terms of near-surface SO2 sensitivity?

    The main goal of the paper is to show that TIR sounders like IASI are able to provide near-surface SO_2 measurements at global scale in case of favorable atmospheric conditions. As highlighted by Reviewer#1 and also mentioned in the paper (introduction and conclusion), the work constitutes a first attempt to retrieve near-surface SO_2 at a global scale using the IASI TIR sounder and provides the first global distribution of near-surface SO_2 from this sounder. Compared to UV/VIS sounders, IASI is able to measure SO_2 during night but also at high latitude (e.g. above Norilsk) during winter. This is already mentioned in the paper in section 3.1 (p11, line 366). Discussing deeper the differences between the capabilities of IASI TIR sounder and UV sounders is more the work of a validation/comparison paper and this is beyond the scope of this work. Following the goal of the work, the present paper focuses more on the capabilities and limitations of IASI measurements only, which are discussed in the whole manuscript as function of TC and H_2O column.

We agree with Reviewer#2 that the advantage of IASI should be highlighted. We have therefore added a sentence in the conclusion, following the addition made in regards to comments of Reviewer#1 (p16, line 535): "*The high-bias is likely linked to overestimation of IASI averages due to the error filtering applied on the data. More comparisons and validation work is needed to investigate deeper the observed differences between the two instruments. The two instruments are nevertheless complementary: regions characterized by high humidity and/or low thermal contrasts can be measured by OMI whereas IASI better monitors SO_2 at high latitudes, especially during the winter, and is not limited to daytime.*"

b. The paper does state that IASI identifies only "dominant anthropogenic hotspot sources". Being a global scale paper it would be really nice to provide a better sense (or quantification) of this statement. Thus, given the general reduced sensitivity of the IR compared with UV/VIS one would assume IASI on a global scale would generally miss a lot of the near surface SO2 sources. Based on the sources that can be seen, what is an estimate of the detection limit both at a single observation level and on a more general emission source level? For example on a emission source level, the UV/VIS OMI instrument is sensitivity to SO2 sources emitting > _ 30 kt/yr (Fioletov et al., 2015), which corresponds to about half of the global anthropogenic emissions of SO2. GOME2 and SCIAMACHY is estimate at 300 kt/yr (Fioletov et al., 2013). Using the sources shown in Figure 6 and contrasting those with Figure 7 of Fioletov et al., (2013), IASI would appear to more closely comparable to GOME2 or SCIAMACHY. Of the 12 sources described in this IASI SO2 paper, all appear to have emissions of at least 600 kt/yr suggesting this might be a reasonable IASI detection limit. Or, since you are already using the EDGAR emissions inventory, it would be very straightforward to compare the sources you are able to detect with value from EDGAR to estimate obtain a detection limit. That said, there are many large 1000+ kt/yr sources that IASI does not appear to able to detect. This should be mentioned explicitly as well. For example, there are many land-based volcanoes in Indonesia that emit over t/yr that are not seen at all, in addition to sources in India and China (as pointed out by the authors).

We understand the question of Reviewer#2 and this would allow having an idea of the capabilities of IASI to measure near-surface SO_2 compared to different UV sounders. However, answering this question is not simple. The sensitivity of IASI is not only limited by the amount of SO_2 released in the atmosphere, but more strongly depends on the values of thermal contrast and H_2O total column. Indeed, even large SO_2 concentrations can be missed by IASI in case of no thermal contrast (according to radiative transfer in TIR) or large H_2O amount (opacity in the near-surface atmosphere in the ν_3 band region due to H_2O absorption, hiding the ν_3 band). Therefore, the detection limit of IASI varies as function of regions but also can vary temporally. For this reason we strongly believe that, unlike as for OMI, the infrared sensitivity to surface SO_2 cannot be expressed unambiguously as a function of emission source level. A figure showing the detection limit (in terms of 0-4 km SO_2 columns) as function of TC and H_2O column is already presented in the paper

(Figure 4). It already shows well the IASI capabilities/limitations in terms of sensitivity as a function of TC and H_2O.
The fact that IASI does not measure sources above India and Indonesia is explained by the large amount of H_2O in these regions, which renders the near-surface atmosphere opaque, i.e. the SO_2 ν_3 band is undetectable. Based on [Fioletov et al., 2013], the smallest source detected by IASI in the global distribution is Sar Cheshmeh in Iran (450±90 kT/year).

In the manuscript, we have added clarifications in section 3.1, p11 last paragraph: "*Comparison with EDGAR database has allowed identifying observed SO_2 plumes. […] Sources in India and in South Eastern Asia are also not observed by IASI. This is likely because of large H_2O amount in the atmosphere in the tropical region, which renders the near-surface atmosphere opaque in the ν_3 band.*"

c. The near surface IASI SO2 product is for a 0-4km layer. For air quality monitoring there is an important difference between being sensitive at 4-km and near the bottom of the boundary layer. It would be good to show from where in this layer the measurement information is coming. One suggestion might be to add a contour plot of the computed Jacobians as a function of height and wavenumber.

We agree with Reviewer#2 that we did not discuss the vertical sensitivity of IASI between 0 and 4 km.. The values of Jacobians (K) as function height can indeed give some insight on the vertical sensitivity between 0 and 4 km. It is important to note that this vertical sensitivity is dependent on thermal contrast values and H_2O amount. Indeed, if there is no thermal contrast and/or large H_2O amount, IASI will be more sensitive to the upper part of the 0-4 km layer. Large TC and dry conditions will increase sensitivity close to the surface. We present in Figure 1 below the Jacobians (from 1 km to 4 km) calculated for different conditions of TC and H_2O total columns.
For the three different conditions presented in Figure 1, Jacobians are the largest at 4 km. With increasing TC, we can see that K at 1 km increases: for TC=10K, the ratio between K at 4 km and K at 1 km is around 3; it becomes 2 for TC=20K. Moreover, for a large H_2O column (left), the K at 1 km are smaller by factor 8 than at 4 km. This is explained by the strong absorption of H_2O which reduces the IASI sensitivity to the atmosphere close to the surface. This also impacts the shape of the Jacobians, particularly in the R-branch. All these results show that, in case of favorable conditions of TC and H_2O, IASI is sensitive down to the surface but has its maximum sensitivity in the upper part of the 0-4 km layer. In case of low TC and/or large column of H_2O, IASI becomes insensitive to the lower part of the 0-4 km layer.
We also refer Reviewer#2 to the answer of the main comment 1 of Reviewer#1 (Figure 1) where we discuss the sensitivity of IASI to SO_2 located in the 0-1 km layer as function of TC and H_2O.

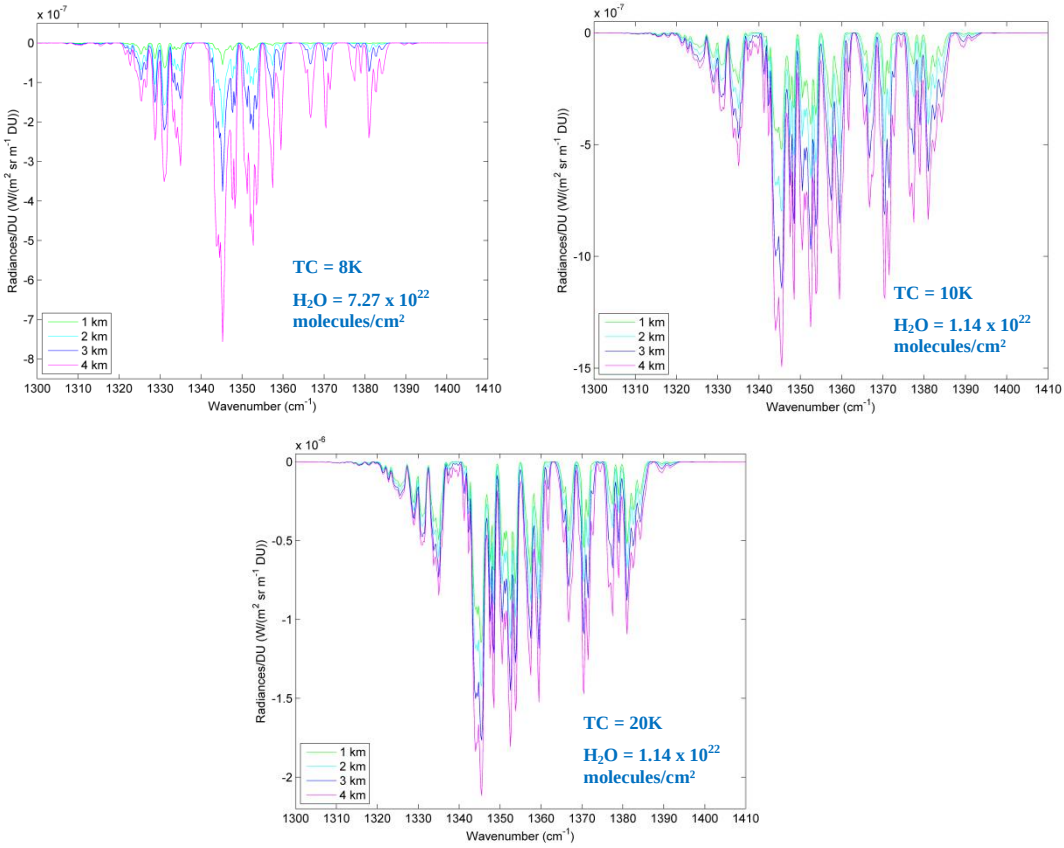

Figure 1 : Jacobians (W/(m² sr m^{-1} DU)) calculated for the ν_3 band of SO_2 for 4 levels (1km, 2 km, 3 km and 4 km). They are presented for different conditions of TC and H_2O total columns.

In the manuscript, we have added a paragraph on this discussion at the end of section 2.2 (p5): "*From the calculated* $\boldsymbol{K_h}$*, we can estimate where in the retrieved 0-4 km layer IASI is the most sensitive to* SO_2*. We found that, for favorable conditions of thermal contrast and humidity, IASI is sensitive down to the surface but has its maximum sensitivity in the upper part of the 0-4 km layer. In case of low TC and/or large column of* H_2O*, IASI becomes insensitive to the lower part of the 0-4 km layer*."

d. As this is a global scale analysis another metric to help gauge the sensitivity of IASI to near surface SO2 globally would be to provide the percentage of valid retrievals relative to the total number of global IASI observations from 2008-2014 (shown in Figure 6). Similar to what was shown in Figure 9 over China.

As discussed in comment 1b, the sensitivity of IASI to near-surface SO_2 depends mostly on TC and H_2O amount. In the paper, we have considered as valid retrievals those associated to less than 25% relative error and 10 DU absolute error (for which we consider that IASI is sensitive to near-surface SO_2). As explained in the answer to the first referee report, this choice favors the largest SO_2 columns. Hence, the percentage of valid retrievals will be heavily determined by the magnitude of the sources, rather than the retrieval algorithm or sensitivity, and would give a wrong message to the reader. For this reason we decided not to show the percentage of valid retrievals but the absolute number of successful retrievals per grid cell for the global

map. This absolute number is still very much influenced by the magnitude of the sources, but in our opinion caries more useful information.

e. Again, the main focus of the paper is global scale near-surface SO2 from IASI. However, the presented detailed comparisons are shown over the highest global anthropogenic SO2 concentration region. To determine the utility of IASI SO2 observations globally it would be good to see the results presented more globally over many regions. For example, since both OMI (likely the current "gold standard" for global SO2 observations) and IASI provide global observations of SO2, why are there not more comparison performed from around the globe under varying conditions. For example, why not also perform the comparison over the Balkhash region shown in the paper?

We agree with Reviewer#2 that the comparison with OMI has been performed above one of the largest anthropogenic source of SO_2. The goal of the paper is the presentation of a retrieval method that allows retrieving near-surface SO_2 from IASI observations. The presented comparison is a preliminary assessment of the method. The Beijing region has been chosen because it experiences seasonal variations of TC and humidity, and allows a comparison in a quite wide range of conditions. Moreover, a second assessment is given by a comparison with results of an iterative method performed on IASI observations recorded above Norilsk. These results were already compared to OMI retrievals and showed a good agreement between the 2 sounders (Theys et al., 2015). As mentioned in the manuscript, we agree that more comparisons with UV sounders but also with in-situ measurements are required to properly validate the retrieval method. This is more highlighted now in the conclusion (see answers to comment 4 of Reviewer#1).
Conclusion: "*The high-bias is likely linked to overestimation of IASI averages due to the error filtering applied on the data. More comparisons and validation work is needed to investigate deeper the observed differences between the two instruments.*"

f. Due to the large high bias in the IASI observations due to its apparent detection limit, the IASI average values are not very representative. One suggestion would be to show a regional spatial map of both OMI and IASI data over the China and Bakhash regions. This would provide the readers with a better sense of the IASI retrievals relative to OMI.

We agree with Reviewer#2 that showing a regional map above China of OMI and IASI can help the comparison. However, please note that these regional averages suffer from the same high bias as the global distributions.

We have included such a figure below the time series of Figure 11. We have modified the text and the caption.
Text (p15): "*[…]Figure 11 shows the comparison in terms of (top) a time series of the monthly averaged columns of IASI (blue) and OMI (red squares for the standard retrieval and green triangles for the retrieval using a different air mass factor (AMF)), and in terms of regional maps (2010-2013) above China (bottom).*"

Caption: *"(bottom) Spatial distributions of SO_2 (DU, different colorscales) above China for the period 2010-2013 from IASI observations (left) and from OMI observations (right, without AMF changes). SO_2 columns have been averaged on a 0.5° x 0.5° grid."*

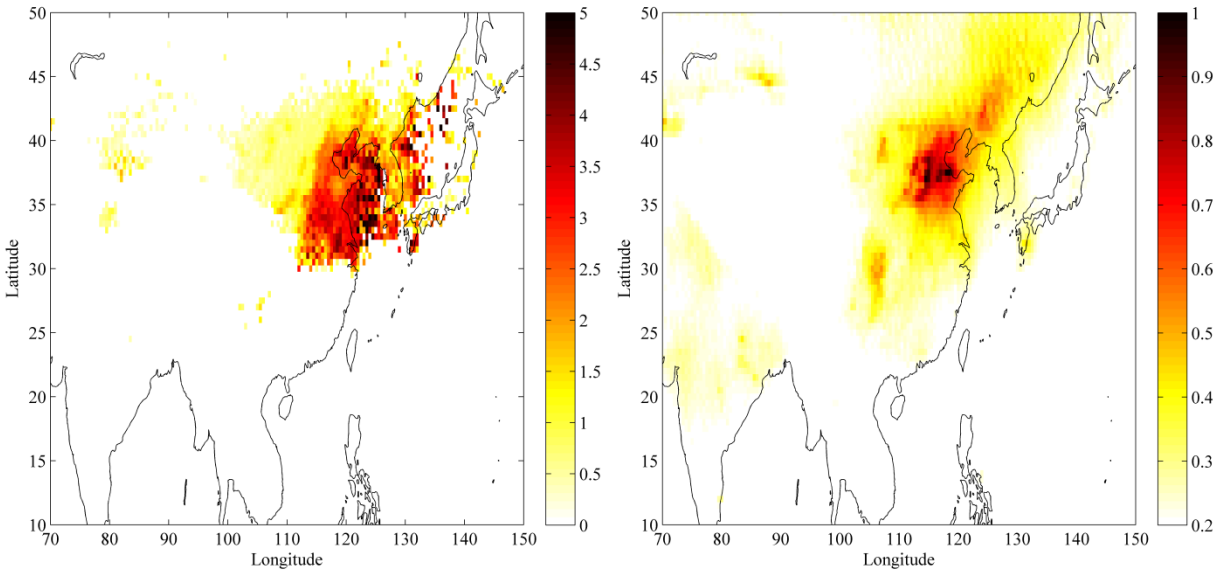

Figure 3 : Spatial distributions of SO_2 (DU, different colorscales) above China for the period 2010-2013 from IASI observations (left) and from OMI observations (right). SO_2 columns have been averaged on a 0.5° x 0.5° grid.

g. Page 11, lines 362-366. Along similar lines to the previous comment, according to Krotkov et al., 2015 there are many moderate to large SO2 sources seen by OMI in these regions (e.g. Figures 6 and 7), which is not consistent with the claims made it this statement. In contrast IASI sees virtually none of these sources (e.g., none in India). Maybe there is good agreement in the global distribution of some/most of the very large emission sources, but this should be clearly stated. The way it is currently written it gives the impression that both instruments have similar SO2 measurement capabilities, which is not the case.

We agree with Reviewer#2 that in both Theys et al. (2015) (Figures 6 and 7) and Krotkov et al. (2015) (Figure 1 and 6), small sources are detectable by OMI above India. However, none of these papers show detected sources above South East Asia (e.g. Thailand), which are reported in the EDGAR database. It is already clearly stated that IASI does not detect SO_2 above India, South East Asia, but also above USA and Europe, and it is explained by small thermal contrast and high humidity (p11, lines 356-360; see also answer of comment 1b). However we have now revised the relevant paragraph to make it even more clear that OMI and IASI do not have the same sensitivity.

The manuscript has been modified at p11, from line 360 to line 368: "*[...] is not limited to IASI. OMI SO_2 distributions (Figures 6, 7 in Theys et al., 2015, and Figures*

1, 6 in Krotkov et al., 2015) shows the ability of the sounder to measure small sources above India, USA and Europe which are not detected by IASI. These distributions also reveal the absence of some of them, compared to those reported by the EDGAR database: South Eastern Asia (e.g. Thailand), Northern Europe and part of India. […] as well as smaller ones, like […] low thermal contrast, undetectable by IASI, can be measured by OMI whereas […].

h. Related to comment 1c), the IR and UV/VIS vertical sensitivity to total column SO2 is different. How might this difference in vertical sensitivity contribute to the differences seen in the IASI/OMI comparisons?

When we look at the 0-4 km range, both OMI and IASI will have their largest sensitivity in the upper part of this range. However in general the decrease in sensitivity for the lowest layers is less drastic for UV/VIS ([Theys et al,, 2013], Figure 1.) . It is therefore correct that the difference in vertical sensitivity in the 0-4 km range will contribute to IASI/OMI differences. This is also interlinked with the fact that the current algorithm assumes a fixed SO_2 profile. Following also comment of Reviewer#1, we have made an estimation of this error (around 30% if SO_2 is confined in the 0-1 km layer for a TC=10K and a total column of $H_2O=9.5\times10^{21}$ molecules/cm²). A paragraph has been added at the end of section 2.3.3 (p8): "*Another source of errors, which is not taken into account in the error calculation, is the assumed SO_2 vertical profile. A same amount of SO_2 but located at different altitudes is indeed associated to different value of HRI. For instance, we have estimated the error on the SO_2 column to be of the order of 30% if SO_2 is confined in the 0-1 km layer (considering a TC of 10K and a total column of H_2O of 9.5×10^{21} molecules/cm²). Note also that the assume temperature profile can be a source of error (see section 3.3).*"
We have also mentioned at the end of p15 (section 3.4.2) that the difference of vertical sensitivity between the 2 instrument can contribute to the observed differences: "*Discrepancies are within the range of what we can expect given the difference in the overpass times of the two satellites and given the high bias introduced by averaging only the IASI observations with a low relative uncertainty. Note also that the difference between the vertical sensitivity profiles of the two instruments can also contribute to observed differences.*"

i. Pg. 12 lines 394: "It is clearly seen in Figure 8 that IASI is mostly not sensitive to surface SO2. . ." Maybe better to use "inferred from" instead of "clearly seen" as no information is provided in Figure 8 indicating where IASI is sensitive.

We agree with Reviewer#2 and we have made the change.

2) Section 3.4.1 on the LUT comparison with an optimal estimation retrieval scheme. Is this section needed? I do see some motivation for comparing a new method with a specific previous one. Provided are the differences between the LUT stated in this paper and the specific OE as implemented and reported by Bauduin et al., 2014. However, to make general statements on the LUT providing better results than OE for low SO2 signals

would require more in-depth information. Fundamentally for any SO2 retrieval method the measurement information from the satellite is coming from the same spectra, thus in theory the physical optimal estimation retrieval should be able to provide the same information/sensitivity as LUT. Thus, it is likely that the differences between the methodologies can be attributed to the specific user selected input/assumptions rather than general methodology differences. If the LUT method shows measurement sensitivity under higher water vapour loading conditions, then a well-designed OE retrieval state should also provide this information and not fall back to the apriori. For example, maybe the OE retrieval is over constrained, etc.? One way to help provide additional insight would be to show the resulting spectral residuals for these cases before and after they have been minimized by the OE retrieval. This would show that: (i) there was information in the spectra to begin with, and (ii) the retrievals reduced them down to the noise level.

We do think that this section is needed. The retrievals performed using an OE scheme in Bauduin et al. (2014) have already been compared to OMI retrievals in Theys et al. (2015). This comparison showed a good agreement between the two sounders. The comparison between LUT retrievals and OE retrievals of Bauduin et al. (2014) provides therefore a preliminary assessment that the LUT-approach is behaving as expected. However, we agree with Reviewer#2 that making general statements on the capabilities of the two methods requires more in-depth analysis. This has also been mentioned by Reviewer#1. The difference observed for high humidity cases is probably related to the fact that there is low signal, which is differently fitted. Therefore, we have changed the end of the section to: "*These measurements have a significant HRI around 5, indicating small signal strength, which is probably the reason of the difference observed between the two methods.*"

3) Page 14, lines 460. less than 20-25% cloud fraction. The cloud fraction is not likely a great indicator in-and-of-itself for species with limited information, the cloud optical depth is more important. For example, 25% pixel fraction from a cumulus cloud might have a large impact on SO2 retrieval. There will also be a dependence on the height of the clouds. Have sensitivity tests been performed to show that a 25% cloud fraction is a good assumption for global IASI SO2 retrievals that have limited information?

We agree with Reviewer#2 , however cloud optical depth is not readily available as an IASI product, and we therefore decided to use this condition which is widely used in IASI-TIR community to select almost clear-sky spectra e.g. see Hurtmans et al. (2012) and Van Damme et al. (2014).

4) Pg. 14, line 464-466. For the algorithm comparison it appears the cases selected for the comparison were based on the quality of the LUT retrievals. Does one get difference results if the cases were selected based on quality OE retrievals instead?

We have selected cases based on the quality of both methods. The selection of observations with "*less than 25% cloud fraction, with a thermal contrast larger than 5 K in absolute value and with a humidity below 4 g/kg at 350 m above ground*" is done for

measurements of Bauduin et al. (2014). These conditions allow selecting observations for which near-surface is probed (p14, line460, and see Bauduin et al. (2014)). For LUT-based retrievals, this selection is made using the errors.

According to the comment of Reviewer#2, this is maybe unclear in the manuscript. We have changed the text of p14, lines 456-459: "*[…] For the comparison, we consider observations located in a circle of 150 km radius around the city of Norilsk. Only measurements of Bauduin et al. (2014) with less than 25\% cloud fraction, with a thermal contrast larger than 5 K in absolute value and with a humidity below 4 g/kg at 350 m above ground have been selected (this altitude corresponds to the average height of the temperature inversions). […]*".

5) I am just curious if it would be more precise to attribute the water vapour impacts on the SO2 retrievals to the water vapour continuum rather than just humidity amounts? I would think that it is not likely the interfering water vapour lines, but rather the more broad water vapour continuum that is of more concern from a radiometric perspective for the SO2 retrievals.

In the spectral range considered for SO_2 retrievals, the H_2O continuum has very small contribution to the absorption as seen in Figure 4. Opacity is mostly caused due to H_2O absorption lines and related to the amount of H_2O in the near-surface atmosphere.

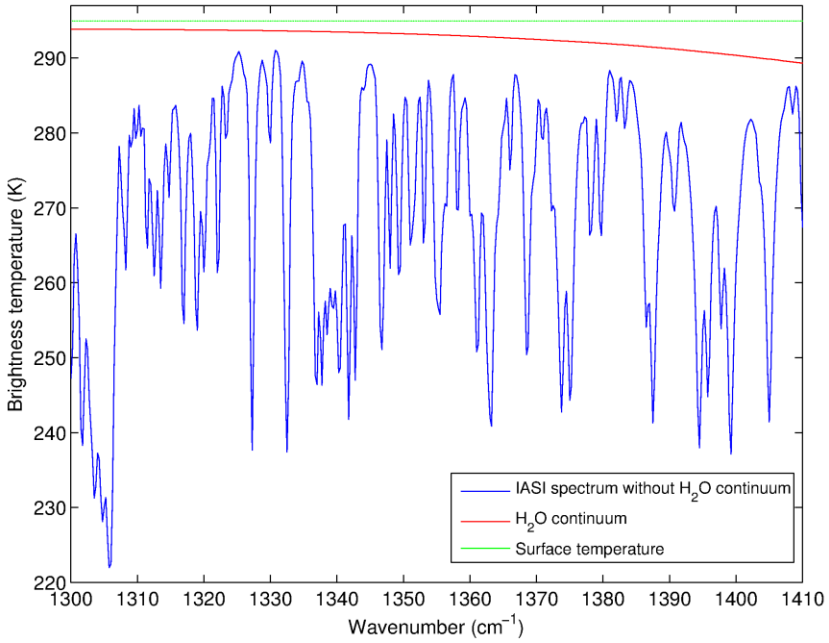

Figure 4 : Contributions of H_2O absorption lines (blue) and contribution of the continuum (red) to IASI spectra. The surface temperature is indicated in green.

6) Section 2.3.2: line 193: In the previous response it was mentioned that comparing a constant and spectral varying emissivity for specified regions are on the order of 0.1K (noise level of the instrument). Since this is a global retrieval, and infrared surface emissivity can vary significantly from the assumed 0.98 value for various surface types, please provide more details on how this is handled in the retrievals. For example, what is the impact of assuming a constant emissivity of 0.98 over different surface types (i.e. desert, water, snow (in winter), etc.)? a. On a related note, on page 11, line 340-345 there is a hot spot 13 over the desert in China. Could this a result of difference in assumed surface emissivity? Why would the plumes only be visible over the desert and not up/downwind of the desert? There is no good reason why OMI would not see this, as OMI is able to see SO2 over the desert (see Figure 7 from Krotkov et al., 2015).

In the spectral range under consideration, the emissivity, even on a global scale does not differ greatly from the 0.98 value (for the ν_1 band of SO_2 this would be a much larger problem) – please see the figures that we posted in our reply to your comment in the quick review for AMTD. Following your comment we now had a closer look at the emissivity over the Taklamakn desert. The maximum difference for the period April-November are of the order of 0.4 K (at a surface temperature of 300 K) if we assume spectrally dependent emissivity. In terms of HRI, based on forward simulations only we found that this could cause a bias of 1.2, which is below the maximum values detected over the desert. Also, the maximum difference (July-August) does not coincide in time with the detection of the plume (March and April). And furthermore, we do not observe any other false identifications over other deserts. For these reasons, we are very hesitant to attribute these observed plumes to surface emissivity anomalies. However, following this comment, we have rephrased the discussion of p11 on the plume (13). It now reads: "*[…] The very high thermal contrast (up to 20 K) and very low humidity conditions found jointly in that region make it indeed possible to measure such weak columns. However, further investigations are still required to properly assess the source of this detected plume and to exclude possible false attribution due to surface emissivity effects."*
We have also changed the text about the description of emissivity in the retrievals (end of p6): "*Note also that a constant emissivity of 0.98 has been used in the forward simulations; for most of cases, differences between using a spectrally varying emissivity and a constant emissivity are the order of the noise of the instrument.*"

7) It would be good to provide the correlation coefficient for the IASI/OMI time series in Figure 11.

The correlation coefficient has been calculated using the reduced major axis method for both set of OMI data (varying AMF and AMF=0.4). It is very low for both cases (respectively -0.03 and 0.15) with a small increase observed with the AMF=0.4. These low values are mainly caused by one outlier on 05/2012, where IASI has a large SO_2 column of 5.7 DU, where there only very few measurements (3). These coefficients are significantly better when this outlier is removed; they become respectively 0.23 and 0.54.
We have now added this coefficient in the text (p15, end of section): "*Finally, we have calculated the correlation coefficient between IASI and OMI measurements. It is very low for both OMI data sets, respectively -0.03 and 0.15 for the varying AMF and AMF=0.4.*

These low values are mainly caused by one outlier on 05/2012, (where IASI has a large SO_2 column of 5.7 DU, which corresponds however to very few measurements (3). These coefficients significantly improve when this outlier is removed; they become respectively 0.23 and 0.54."

8) Page 16, lines 543-545. To be complete in terms of the trade-offs between the two retrieval approaches some of the main benefits of a OE retrievals should also be noted. For example, robust straight-forward error estimates, direct method for accounting for apriori information in comparisons, sensitivity and information content provided for each retrieval via averaging kernels, straight-forward computation of observation operators for the direct inclusion of satellite retrievals in chemical model assimilations/ inversions, etc.

Following comments of Reviewer#1, we have removed the sentence "*It is also very sensitive and has shown interestingly better results for weak SO_2 signals*".
We have also added advantages of the OE method: "*[…] It has the advantage of being very fast; iterations and the retrieval of interfering parameters are not needed. In contrast to optimal estimation method, it is however not able to provide a full retrievals characterization of the retrieved columns/profiles, notably in terms of vertical sensitivity.*"

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