Response to Reviewer #1

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.

In the manuscript the authors present a study of 'near-surface SO2' based on the IASI measurements, using the IASI spectra around v3 absorption band. This is the first global map of low level SO2 from IASI and I suggest it for publication. The work is original, contains important addition to existing literature and is in general well written. Nerveless it will need some clarifications and additions as my following comments.

Main comments:

1) The use of only the v3 band to study close to the surface SO2 is not the best exploitation of IASI spectra. The present work is still valuable and I suggest it for publication but the presence of V1 band within IASI spectra have to be empathize more. I can accept there are practical reason for using only the v3 band, (as for example missing a good forward model, surface temperature/thermal contrast problem etc...). The v3 absorption band is within the water vapor absorption band, and it is in general not sensitive up to the surface and this should be mentioned (at the end of the introduction and in the conclusion). A work using both band v3 and v1 is desirable and it will carried more information on SO2 close to the surface that using only v3 band. The fact that they use only the v3 band should be mention in the abstract. I really don't won't that the reader to have the impression that IASI cannot measure SO2 close to the surface in condition with a lot of water vapour, this is a limitation of this retrieval not of the IASI measurement itself.

   We agree with Reviewer#1 that the ν_3 band is affected by the strong absorption of water vapor, which can make the near-surface atmosphere completely opaque in this spectral range. The ν_1 band is indeed less impacted by H_2O absorption and can provide valuable information about surface SO_2. However, the ν_1 band is weaker than the ν_3 band by about a factor 7.8 if we compare the strongest lines of each band [Bauduin et al., 2014]. Therefore, whether the ν_3 or ν_1 band will bring more sensitivity to near-surface SO_2 very much depends on the atmospheric conditions.

   We explored this further with the help of Figure 1 below, which represents the spectral signal detected by the IASI instrument (color bar) due to an enhancement of SO_2 of 100 ppb well-mixed in the 0-1 km layer for an atmosphere representative of mid-latitudes in winter (left) and summer (right). It has been calculated by the difference between a simulated spectrum with 100 ppb SO_2 well-mixed in the 0-1 km layer (US Standard profile above) and a simulated IASI spectrum with only the US standard SO_2 profile. These differences have been calculated for a large range of thermal contrasts. They have been normalized by the noise of the instrument; a normalized difference larger than one is thus needed to be able to see the enhancement of SO_2 with IASI. The total column of H_2O is 2.86×10^{22} molecules/cm² and 9.75×10^{22} molecules/cm² respectively for winter and summer. The 0-1 km column of SO_2 is of 8.81 DU for simulations with enhancement of SO_2; when there is no enhancement, the 0-1 km column of SO_2 is of 0.03 DU. From this figure, two important comments can be made:

- For dry conditions (winter), IASI is able to detect the enhancement of SO_2 in the 0-1 km in the ν_3 band from around 3K of thermal contrast (absolute value). With this result, it clearly appears that IASI is sensitive to SO_2 down to the surface in the ν_3 band for dry conditions. This is not the case for larger amount of H_2O (summer), in which SO_2 signal is undetectable in the ν_3 band. These results are in the line of those presented in the paper, in Figure 4 (detection limit) and in Figure 8 (time series).
- As already mentioned, the amount of H_2O has less impact on the ν_1 band. However, even for the large column of SO_2 considered here, this band is only weakly detected, even for large thermal contrasts. Thus the ν_1 can bring information, but only for very large concentrations.

Because of the weakness of the ν_1 band, we chose to use the ν_3 band for the retrieval. We agree with Reviewer#1 that the use of both bands could improve the results and this can be envisaged in the future development of the product. We also refer Reviewer#1 to the comment 1-h of Reviewer#2 about vertical sensitivity.

To clarify this in the manuscript, we have changed the end of the introduction (from line 27 of p 11032) for: "*This calculation is performed using the ν_3 band (1300-1410 cm^{-1}). This spectral band has the advantage of being more intense than the ν_1 band (factor 7.8 larger for the strongest lines, Bauduin et al., 2014). It is however more affected by the absorption of H_2O, which can cause opacity in this spectral region in the near-surface atmosphere and therefore can reduce the IASI sensitivity to SO_2 down to the surface. This parameter, along with thermal contrast, has been taken into account in the whole retrieval procedure (described in section 2) and the results are analyzed with respect to these two parameters*."
We have also changed the conclusion (p11053, lines 14-15): "*The use of both ν_3 and ν_1 bands can also be envisaged to reduce the impact of H_2O absorption and to increase the IASI sensitivity to surface SO_2.*
Note that the use of the ν_3 band was already mentioned in the abstract (line 11) and has now been mentioned in line 20.

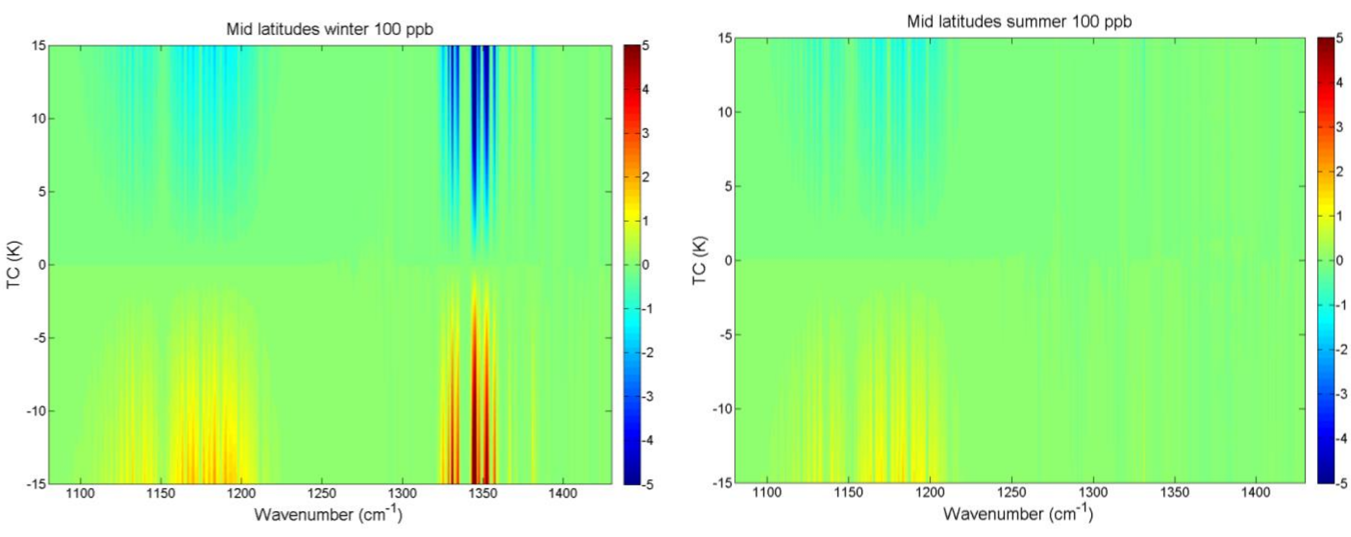

Figure 1: Differences between a simulated IASI spectrum with 100 ppb SO_2 well-mixed in a 0-1 km layer (US Standard profile above) and a simulated IASI spectrum with only the US standard SO_2 profile. The differences are normalized by the instrument noise (color bar) and calculated for different values of thermal contrasts. Atmospheres representative of winter and summer conditions for the mid-latitudes [Anderson et al., 1986] have been used (left and right respectively).

2) This work is missing the sensitivity of the linear detection HRI(h), how much SO2 do you need to 'detect' something between 0-4 km? How much if SO2 is between 0-4 and how much if SO2 is between 0-1 km? I'm not sure if they are using the HRI(h) to reject the pixels with h > 4 km or if they are using HRI(h) select the pixels with h < 4 km, please clarify this somewhere in section 2.

The altitude retrieval was used to select the pixels with an h<4 km. An altitude was retrieved when a HRI larger than 2 was found, even though in practice we expect the altitude retrieval to be accurate only for HRI values above 4 or 5. In terms of SO_2, the amount greatly depends on the thermal contrast and total column of H_2O, and follows the same trend as in Figure 4 (for the 0-4 km).

Note that the exact SO_2 limits for which the altitude retrieval can be trusted is not relevant in the current retrieval scheme, as when the altitude retrieval is not made (HRI<2) or unreliable, the observation is unlikely to pass the strict quality flags on the retrieval uncertainty. The main goal of the altitude retrieval here is removing SO_2 from large high altitude volcanic eruptions.

We have now added the HRI threshold of two to the manuscript (in section 2.2 on p11036, line 4): "*Note that an altitude was retrieved when an HRI larger than 2 was found, even though in practice we expect the altitude retrieval to be accurate only for HRI values above 4 or 5.*"

3) The data cut implemented a posteriori really affect the global map presented and all the results are biased high, in particular they are averaging only values bigger than some 'value' and presumably are sensitive to only SO2 higher than some 'altitude' (I do think that the work is still valuable and that v3 band is sensitive below 4km, but not sure it is sensitive up to the surface). It will be a nice addition to estimate these 'value' and altitude for a specific atmosphere (standard atmosphere?).

The comment on the high bias will be addressed in the reply of the next comment. For the sensitivity up to the surface, please see the reply above. Here we answer the question about how the posteriori flag translates in terms of SO_2 concentrations on the 0 to 4 km range. For this, we have estimated the 0-4 km column of SO_2 above which the relative error will be smaller than 25% for different conditions of TC and H_2O total column in the table below:

Table 2: 0-4 km column of SO_2 (DU) above which the relative error is smaller than 25% for different conditions of TC and H_2O total column

	TC=3K	**TC=10K**	**TC=15K**
$H_2O=5.3\times10^{21}$ molecules/cm²	3.3 DU	0.8 DU	0.7 DU
$H_2O=2.01\times10^{22}$ molecules/cm²	4.7 DU	2.2 DU	1.0 DU
$H_2O=5.05\times10^{22}$ molecules/cm²	14.5 DU	10.8 DU	9.9 DU

4) You apply a posteriori cut for cloud cover and for SO2 errors. I agree with the cut for cloud fraction (e.g. if there is cloud IASI don't see the surface), but I think that the cut you are doing for error in SO2 load (< 25% and <10 DU) is introducing a sampling bias and really affect your global map. Values which are zero should still contribute to the calculation of global mean distribution (and cut values with error >25% is equivalent to cut low values). What happen if you cut only for <10 DU and accept any relative error? It will 'save' a lot of pixels with low amount of SO2 to go into the global distribution. I think that applying these cuts (= accepting only < 25% and <10DU) is equivalent to consider only SO2 in some defined range of amount and altitude. Apply a relative error cut means that only the retrieval above some absolute amount of SO2 (which depend on altitude) will contribute to the mean. To understand how the SO2 distribution if Fig.6 is limited by the actual presence of SO2 or by sensitivity of the algorithm, it will be informative to add a map illustrating the sensitivity, e.g. the mean absolute error or the percent of retrievals that have error <10DU. Why you don't you do an errorweighted average without any cut based on the error? Given that approach from Fig.6 is not possible to judge the significant of the blue values plotted especially without any indication of the percent of pixels that have gone into the "average" – the map looks reasonable but blue values could be generated by single retrievals randomly crossing the quality control thresholds.

We agree that the error posterior filtering introduces a bias in the presented averages. The chosen error filtering allows rejecting measurements for which IASI sensitivity to near-surface SO_2 is limited and thus for which associated retrieved SO_2 columns should not be trusted. The efficiency of this filter is notably proven with the time series made above Beijing (section 3.2), where periods with no SO_2 measurements correspond to periods with low TC and/or high H_2O total column. The condition absolute error>10DU rejects only few points corresponding to unrealistic large SO_2. Using weighted averages (on relative error) would also have favored the larger concentrations [Van Damme et al., 2014]. It is important to stress that averaging data for which the measurements sensitivity is so variable will however, always introduce biases. Averages (maps) should therefore be interpreted correctly; they are averages of those measurements that pass the quality criteria, and not necessarily approximate averages of the true SO_2 columns.

In the manuscript, we have added a paragraph at the beginning of section 3 (p11041, line 8) to explain to the reader that averages are necessarily biased high: "*In this section, global distributions, time series and a first product evaluation are presented. For this, only SO_2 columns with less than 25% relative error and less than 10 DU absolute error have been used. The second criterion was necessary to remove spurious data over the cold Antarctic region with unrealistic high columns. The first condition has been chosen to reject measurements for which IASI sensitivity to near-surface SO_2 is limited, and thus for which associated retrieved SO_2 columns have large uncertainties. However, this procedure tends to favor large SO_2 columns. As a consequence, the presented averages are expected to be biased high. It is important to stress though that individual measurements that pass the filter are not a priori biased but have random uncertainties related to errors on the different input parameters (errors on TC, choice of temperature and SO_2 profile).*"

After this new paragraph, following the comments of the reviewer on the observations with SO_2 close to 0, we have added the following (p11041): *"It is important to note that, by also using the HRI>2 criterion early in the retrieval procedure, we have made the choice not to treat observations with small or undetectable amounts of SO_2. This potentially throws away one category of useful observations: those where a low HRI is found together with favorable atmospheric conditions. In this case, a low HRI can be an indication of the absence of large SO_2 concentrations at the surface. Future versions of the retrieval algorithm could be expanded to include those, and this would potentially decrease the high bias when making averages."*
Following the comment of the referee we have now also added in Figure 6 an inset with the total number of successful measurements in each grid box.

In the sub-sections, we have changed the sentences referring to the error filtering (p11041-lines 15-18, p11046-line 15, p11049-lines 24-25) as everything is now explained at the beginning of the section.

We have added a remark in the discussion about comparisons with OMI (p11051, line 18): "*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.*

Finally, we have added a comment on this in the conclusions (p11052, line 24): "*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.*"

Specific comments:

- p 11032 l 28- p 11033 l 1: why you are not using v1 band? You should mention that v1 band is desirable for studying values close to the surface.

  This comment has been addressed in the main comment 1.

- p 11035 l 8,9: '. . . calculating the function HRI(h) and finding the altitude of its maximum' to change into: '. . . calculating the function HRI(h) at predefined altitudes, and finding the altitude of its maximum'.

  We agree with the suggestion of Reviewer#1 and we have made the change.

- p 11036 l 8-11. It is not clear how they use the HRI(h): (a) to filter out the pixel that result with detection of SO2 > 4 km; or (b) they use only the pixels that result positive to a detection between 0-4 km?

  This comment has been addressed in the main comment 2.

- p11040 l3-5 If the answer above is (2) then this sentence is not true and they have to check the detection limit of HRI(h), when the detection 0-4 is positive and when it is not?

  Please see the answer to the main comment 2.
  We do not fully understand the remark of Reviewer#1. The calculation of the detection limit relies on the LUT and is not related to the function HRI(h). For the LUT, one HRI is calculated for each simulated spectrum, corresponding to the spectral signal associated to the chosen 0-4 km column of SO_2 (and profile). The limit of detection has been set to HRI=3, and the corresponding 0-4 km SO_2 columns has been defined as the detection limit. We therefore decided to keep the sentence as it is.

- p 11040 Error characterization: A big source of error is the SO2 profile assumed. IASI v3 band is not sensitive up to the surface even in dry conditions. IASI signal in v3 is affect by SO2 above some threshold altitude (_1km?). So you are assuming the SO2 amount below that threshold altitude according to your profile. (I agree this threshold depend on water vapour but anyway it is not zero.) It is hard to estimate the error associate to the assumed profile, but it should be mentioned in the paper as source of error.

  We agree that the choice of SO_2 profile is a source of error and that this was not sufficiently highlighted in the manuscript. However, as demonstrated in the answer of the main comment 1, in dry conditions, the signal in the ν_3 band is also sensitive to the 0-1 km layer. 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. We have also added a comment on temperature profile.

  In the paper, a paragraph has been added in section 2.3.3 on error characterization.
  "*Another source of errors, which is not taken into account in the error calculation, is the assumed SO_2 vertical profile. A given column amount of SO_2 located at different altitudes corresponds to different HRI values. For instance, we have estimated the error on the SO_2 column to be of the order of 30% when SO_2 is confined to the 0-1 km layer only (for a TC of 10K and a total column of H_2O of 9.5×10^{21} molecules/cm²). Note also that the assumed temperature profile can be a source of error (see section 3.3).*"
  We have also mentioned the SO_2 profile in the future development described in the conclusion (p11053, line 13): *"[…] of the variety of temperature, SO_2 and H_2O profiles occurring globally*."

- p11041 l5-7: which are the conditions that fulfil the surface sensitivity?

  These conditions are difficult to establish. We estimate that IASI becomes sensitive to 0-4 km SO_2 from TC=3K (in absolute value) and below a H_2O total column of 4×10^{22} molecules/cm². However lines 5-7 were perhaps not very clear; now that we have added comments on error filtering at the beginning of section 3, we removed them. The end of the 2nd paragraph now reads: "*The errors are used in the following to filter out the data (see section 3).*"

- p 11044 l 21-24. If the majority of HRI is below 5, why your fig 2 and fig 5 have y-axes that go up to 1600 HRI?

  The majority of HRI is below 5 for the particular case of the Taklamakan desert. This is not the case for all the sources and the LUT have been built to be able to retrieve SO_2 for extreme cases. In practice the largest HRI encountered was 309.

- p 11045 l 17-19. 'Sources in India and in South Eastern Asia are also not observed by IASI, likely because of large H2O amount in the atmosphere in the tropical region.' These sources are not observable by IASI using the v3 band only, but it may be possible to observe them with the v1 band. You can mention here that the use of v1 band could help.

  We agree with Reviewer#1 that the use of the ν_1 band could help detecting these sources. We have added a sentence at p 11045, line 19: "*Note that the joint use of the ν_1 band, less impacted by H_2O absorption, could allow detecting these sources.*"

- p 11046 l 23 'but the humidity is too high during the other months to allow IASI probing the surface.' to change into: 'but the humidity is too high during the other months to allow probing the surface using this IASI scheme.'

  We have made the change.

- p 11047 l 14-15 I'm not sure I understand what is not covered in the LUT range, do you means that the thermal contrast was less than -30 (as you range of thermal contrast from table 1)? I'm not sure I understand the problem, why you don't extend the LUT range?

  It is not a problem related to the range of TC covered by the LUTs. For some measurements associated to small negative thermal contrast (0>TC>-10), the calculated HRI is more negative than the values allowed by the LUT. As explained in section 2.3.2, the competition between emission and absorption in the first layers of the atmosphere depends on TC but also on the temperature profile. We have found that because we use only one temperature profile to build the LUT, some measurements do not have a correspondence for their HRI value, because our chosen temperature profile do not allow such HRI value for the conditions (TC, H_2O, angle) of the measurement. To correct for this, we have to take into account the variety of temperature profiles, as discussed in section 3.3 and in the conclusion, and this is more complicated to implement and will be the object of future development of the method. Note also that the assumed vertical profile of SO_2 can also be responsible for this no correspondence.
  We have added a small clarification at p11047, line 15 to explain that it is the HRI value that has no correspondence: "*[…] were not covered by the LUTs (i.e. their HRI values have no correspondence in the LUT).*"
  We also now mentioned the vertical profile of SO_2 at p11048, line 17: "*Note that errors on the thermal contrast and on the assumed SO_2 vertical profile also affect the HRI and, as a consequence, the retrieved SO_2 column. They could be partly responsible for the observed non correspondence between measured HRI and the LUTs*".

- p 11048 l 7-8 Again why not simply increase the LUT range? Otherwise you should really document which conditions are outside your analysis, e.g. add a map illustrating the sensitivity as main comment (4).

  This is an illustration of the problem discussed in the previous comment and is inherent to the way the LUTs have been built. This problem occurs mainly for nighttime measurements as mentioned in this section.

- p 11048 l 23-25 'the average are bias high'. This is true not only for the night measurements, it is true also the day and dry measurements, they will be biased high as well. It will be a different bias (a lower bias) but you are still overestimating the average. You are averaging the SO2 only when the IASI v3 is sensitive with little error, so it is bias high anyway. The map in fig 6 are not an 'average' they are a global distribution of detected signal, e.g. signal that overpass some amount.

  As discussed in the main comment 3/4, we agree with Reviewer#1 that even during day, averages are biased high. This is already mentioned in the following lines (25-27) and now, it is also highlighted in beginning of section 3.
  In the text we have modified line 25 and removed "*probably*". We have also added a comment on the discussion of the global distribution (at p11045, line 12) and in the caption of Figure 6: "*[…] like volcanic eruptions. Finally, as mentioned in the beginning of section 3, because of the error filtering, the presented global averages are biased high. It is therefore an average of measurements which are sensitive to near-surface* SO_2*.*"

- p 11050 l 10-17 This paragraph is not 100% clear. I don't think that when there is discrepancies between iterative retrieval and the linear one it means that the linear one is correct. It simply means that when there is low signal the iterative and the linear give different results. Please delete the last sentence 'This result shows the strength of the LUT-approach for these low-signal cases.'

  We agree with Reviewer#1 and we have changed the end of the paragraph (lines 14-16): "*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.*"

- p 11052 l 6 '...the posterior selection of the retrieved SO2 columns for which IASI is sensitive enough.' This is really a key point and should be explained better, please document the minimum values of So2 amount and altitude that you retrieve. For example in a standard atmosphere conditions, applying cut of 25% and 10 DU errors in SO2 amount, which valued can you retrieve? I think this correspond at least in cutting all the data that have _<2 DU and with altitude _<1 km but it will be important to know the proper numbers for this scheme.

  This comment is related to the main comment 4 and has been answered.

- l 14: This is only a suggestion: the Taklamakan desert SO2 source can be a Potassium Chloride facility, as you can see in wikipidia (sorry I don't have better source): https://en.wikipedia.org/wiki/Lop_Nur

  We thank Reviewer#1 for his/her suggestion. Indeed, this could be a possible source. However, we did not find a better reference and we have decided to not add this in the manuscript.

- p 11053 l 4-5 Nothing in this manuscript demonstrate that the linear retrieval is better than the iterative one, please delete this sentence: 'It is also very sensitive and has shown interestingly better results for weak SO2 signals'.

  We agree with Reviewer#1 and we have removed the sentence.

- p 11053 l 8-12. I'm not sure I think that the difference in day/night can also come from the data cut applied.

  A part of the difference comes indeed from the error filtering, which probably rejects more measurements at night than during day because of generally smaller TC encountered during night. This was discussed in section 3.3. However, the most important issue during night is the absence of correspondence in the LUT for some observations (see also answers of previous comments), which results in the rejection of most of the measurements. We agree that in the conclusion, the larger high-bias of night averages is not mentioned and we have added a comment at p11053, line 9: "*[…] photochemistry and a larger high-bias in the night averages could explain part of this effect […]*".

- p 11053 l 14-15 'The use of the v1 band can also be envisaged to reduce the impact of humidity and increase the number of used data.' To change into: 'The use of the v1 band can also be envisaged to reduce the impact of humidity and increase the sensitivity close to the surface.'

  A change has already been made following the main comment 1 and is in agreement with this comment.

- Fig 1: how much is in DU the profile plotted here on the left?

  The profile corresponds to a 0-4 km SO_2 column of 0.4 DU and we have now mentioned it in the caption of Figure 1. We remind Reviewer#1 that this profile has been scaled to take into account different amounts of SO_2.

- fig 2: (a) is mainly all blue, please use a different colorscale. Maybe a logarithmic one? Why are the values going up to 1600 HRI? Maybe it can be a plot in smaller y-axe, and you can zoom more in the interesting part (the central one). Are these HRI, in the range -400 +1600, values that you find in the analysis?

  We agree with Reviewer#1 and we have changed the colorscale in Figure (a) and (b). We however kept the axis as they were to show the totally of the ranges covered by the LUTs and to see the variation of the HRI values as function of TC and H_2O (notably for the discussion of section 2.3.2). In our analysis, the values of HRI are not as large (small) as the maximum (minimum) of shown values. As mentioned in comments above, the largest HRI encountered was 309.

- fig 4: again a lot of blue here, what about a log scale colorbar? Or a non linear or different colorbar. Values between 0 and 3 DU are mainly indistinguishable.

  We have changed the colorscale to better distinguish values between 0 and 3 DU.

- fig 5: it is nearly all blue. Please use different colorscale (log one?). Plus do you really need this range or can you zoom it to something around -10 10 TC and -100 100 HRI? I will prefer this plot in absolute value (and not relative) to go together with fig 2. Maybe you can plot both the relative and the absolute errors?

  We have changed the colorscale of the relative error plot. Moreover we have added a plot with the absolute error. We have modified the caption and the description in section 2.3.3 according to this addition.

- Figure 6: these are average of the data that exceed some threshold values, e.g. the amount of SO2 that i needed to 'activate' the HRI(h) for 0-4 km, and have to be in favorable condition to pass the error thresholds. See my previous comment (4).

  We agree with Reviewer#1 and we refer him/her to the answer of previous comments.

References:

- Anderson, G., Clough, S., Kneizys, F., Chetwynd, J., and E.P., S.: AFGL Atmospheric Constituent Profiles (0-120km), AFGL-TR-86-0110, Environmental Research Papers, 954, ADA175173, 1986.
- Bauduin S. et al. (2014), IASI observations of sulfur dioxide (SO2) in the boundary layer of Norilsk, Journal of Geophysical Research: Atmospheres, 119, 4253-4263, doi:10.1002/2013JD021405.
- Van Damme M. et al. (2014), Global distributions, time series and error characterization of atmospheric ammonia (NH3) from IASI satellite observations, Atmos. Chem. Phys., 14, 2905-2922, doi:10.5194/acp-14-2905-2014.