Answer to the referee 2

First of all, we would like to thank the referee for her/his review of our paper and for giving us the opportunity to improve our paper.

The answer to the comments is organized as follows. First, we list some notations that will be adopted in the answers and the main changes done to the paper. Then, we detail our answers to the questions raised by the referee.

Notations:

- **Old version of the paper**: means the version submitted before.
- **New version of the paper**: means the version we submitted after the modifications based on the referee's comments.
- The '**_R2'** added in the legend of the figures: means referee 2.

Changes:

- We have removed Figure 3, 4 and 5 (of the old version) from the new version of the paper (the reason is detailed in the comment 1).
- We have modified Figure 7 (of the old version) to show data averaged by grid box as suggested by the referee 1 (see comment 23 of general comments of referee 1).
- Table 1 was extended to include other lines.
- RefExp is replaced by RdiagExp in the new version of the paper and in the answers too.

Answer to the questions of the referee:

The question is copied in italic and the answer is written in normal font.

I. General Comments:

1. *I would welcome a discussion on why the separate treatment of the land/sea covariance matrices did not yield significant results. Otherwise I think the land/sea and day/night results could be cut:*

Answer

Since the separate treatment of land/sea covariance matrices did not yield significant results, we propose in this new version of the manuscript to keep only one paragraph discussing this aspect (L15 P10 to L10 P11 of the new version). As suggested in the comment of the referee, we cut the figures of day/night and sea/land discussion (Figure 3, 4 and 5 of the old version of the paper). We give here more details for this choice.

As we pointed out in the first version of the submitted paper, the separation of the type of the surface of observations during the assimilation did not show a significant difference with the case of considering a global estimation.

Figure 1_R2 shows the relative difference of the RMSE with respect to radiosoundings (see the formulation used to compute them in comment 21.2 of specific comments) for an experiment using the R estimated globally (green line), R estimated with separation of the type surface (red line, where each pixel is attached to a matrix estimated according to the type of the its surface (land/sea)), and R diagonal (blue line). The same validation was adopted in figure 9 of the paper (old version). The validation against ozonesondes shows slight differences with small improvements around 150 hPa in the tropics and in the southernvmidlatitudes (30S-60S) free troposphere.

Figure 2_R2 shows the same results reported in the figure 8 of the manuscript (old version): Difference of the ozone total column (DU) provided by OMI and that of the assimilation experiment using an estimated R-matrix globally, averaged over the month of the study.

Figure 3_R2 shows the difference of the ozone total column (DU) provided by OMI and that of the assimilation experiment using an estimated R-matrix according to the type of the surface (land/sea) averaged over the month of the study.

A comparison between Figure 2_R2 and Figure 3_R2 shows that the separation of the type of the observation's surface type did not yield significant differences in terms of total column.

This behavior might be explained by the number of observations over the sea and over the land. In fact, the observations over the sea represent more than 70% of the total of observations. As we can notice in Figure 4_R2, the differences, in terms of standard deviation, of the global estimation and that using pixels over the sea is very small in comparison with that using pixels over the land. The differences are also small in terms of correlations in the case of the sea surface in comparison with the land surface (Figure 5_R2). Hence, we consider that the predominance of observations over sea averages out the potential differences caused by a separate land/sea specification of R. This papragraph was added to the paper (L27 P16 of the naw version of the paper)

As it was suggested by the referee's comment, we removed the figures of day/night and sea/land comparison (Figure 3, 4 and 5 of the old version of the paper) and we keep only one paragraph (L15 P10 to L10 P11 of the new version) that resumes this discussion.

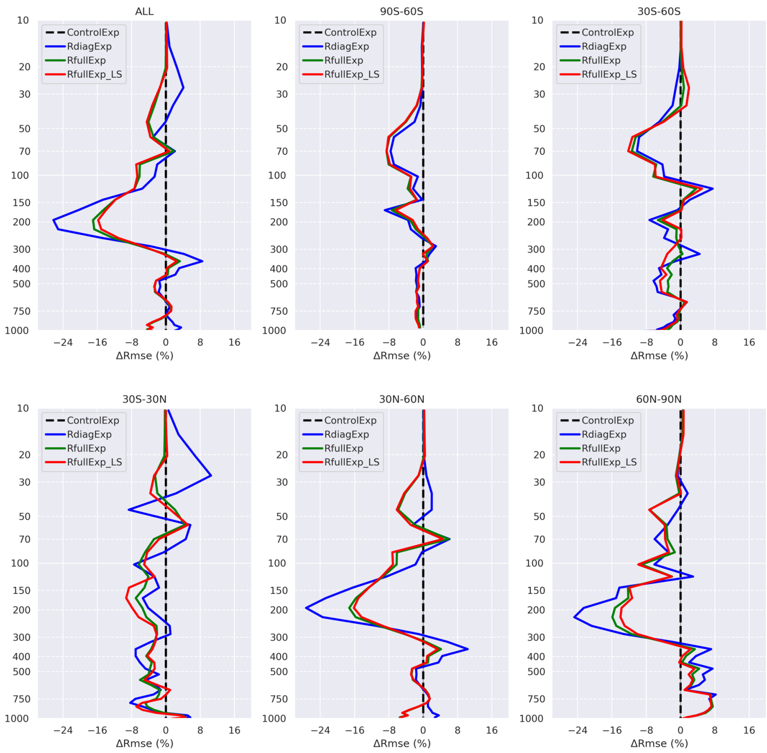

Figure 1_R2: Normalized difference of the RMSE with respect to radiosoundings for the RFullExp (green), RdiagExp (blue) and the RfullExp_LS (the experiment using separated matrix according to the type of the surface) in red. The difference of the RMSE was computed by subtracting the RMSE of the controlExp from the RMSE of the analysis of each experiment, divided by the average profile of radiosoundings (see the formulation in comment 21.2 of specific comments).

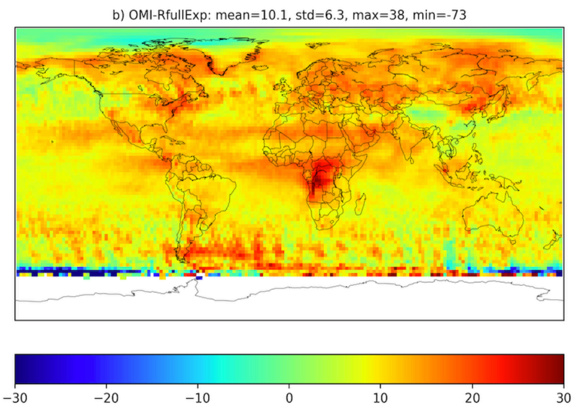

Figure 2_R2: Difference of the ozone total column (DU) provided by OMI and that of the assimilation experiment using a matrix estimated over the glob averaged over the month of the study.

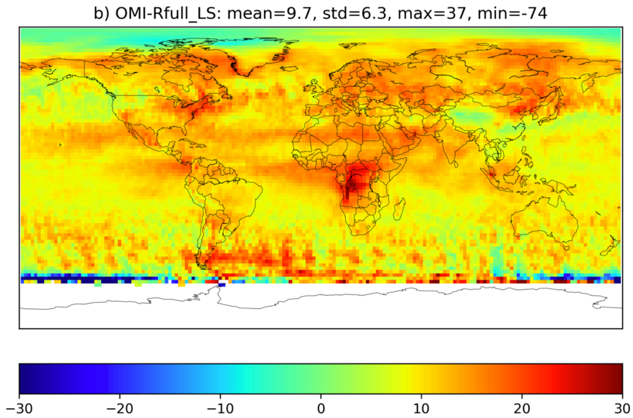

Figure 3_R2: Difference of the ozone total column (DU) provided by OMI and that of the assimilation experiment using a matrix according to the type of the surface (land/sea) averaged over the month of the study.

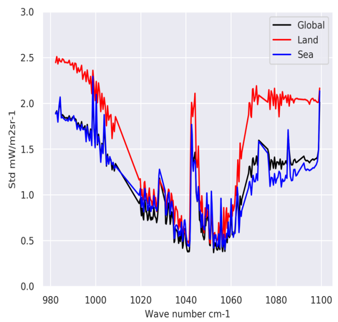

Figure 4_R2: Standard deviation estimated using Desroziers diagnostics according to the type of the surface (sea, land and global).

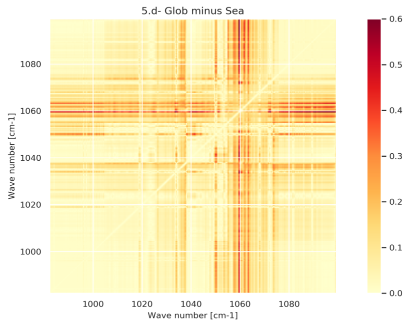

Figure 5_R2: Difference (in %) between global and sea correlation matrix (divided by the global matrix).

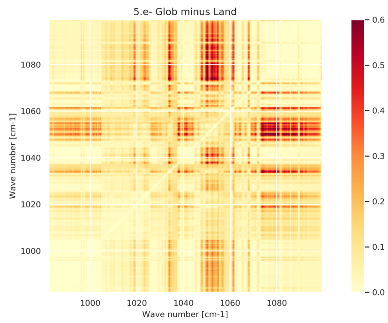

Figure 6_R2: Difference (in %) between global and land correlation matrix (divided by the global matrix).

2. *Condition number discussion*

2.1. It seems that reconditioning was only used to correct negative values which could result in a nearly singular matrix?

Yes, in fact the conditioning method was based on the correction of the negative values. The objective was to obtain a symmetric positive definite matrix. The resulting matrix shows, indeed, very strong interchannel error correlations and remains relatively ill-conditioned. Nonetheless, to ensure that the inversion of the matrix was performed correctly, we computed the product of the R-matrix and its inverse and we checked that it is equal to the identity (with a precision of 10^{-4}), before any further use within the assimilation.

2.2. What is the minimization algorithm? Does it include a preconditioner that depends on R?

The minimization algorithm used in this work is LBFGS (Liu et al., 1989).

No, it does not include a preconditioner that depends on R. But, the system is preconditioned with the square root of the B-matrix.

The paper was modified to include this comment.

2.3. *What are the final condition number?*

The majority of eigenvalues are very small comparing to the maximum value. That makes it a bit difficult to get a well-conditioned matrix without changing dramatically the matrix. We have verified that our matrix was well inverted before any further use within the assimilation. The final condition number is: $8.\ 10^{6}$.

3. *Was bias correction used and if so, what method? If not, were significant biases observed in the IASI observations?*

3.1. *Was bias correction used and if so, what method?*

Answer

No bias correction was used in this study. A detailed discussion is given in the response to R1 and reported here for completeness.

In NWP, the systematic errors in satellite observations are in general corrected before assimilating the observations or within the data assimilation process by VarBC scheme (Auligné et al., 2007). The key assumption is that the background state provided by the NWP system is unbiased. This assumption is not valid in atmospheric chemistry applications, where models might have significant biases, which is the case in our study (see figure 4 in Emili et al., 2019). In such case, VarBC requires some independent data (anchor) to prevent the drift of the analyses to unrealistic values that might be introduced by the model bias. In our case, we control tropospheric and stratospheric ozone. Identifying an anchor needs to be investigated carefully.

Ozonesondes might be used as an anchor in the troposphere and low stratosphere, but the number of profiles provided is limited spatially and temporally. This might have an impact on the capacity of ozonesondes measurements to prevent the drift of the analyses due to the model bias. Han et al. 2010, have used the channel 1585 (9.61μm) as an anchor in the assimilation of ozone for NWP. Dragani et al. 2013, have used the same uncorrected channel as anchor and they showed that its impact was not sufficient to stabilize the bias correction process for the long period. This aspect needs to be explored carefully in a separate study.

On the other side, a good understanding of sources of the measurements bias is a prerequisite to implement a bias correction scheme. VarBC in NWP applications, for instance, needs to define a linear model with some predictors (Auligné et al., 2007). Before adapting this approach in atmospheric chemistry framework, the possible sources of systematic errors in IASI ozone window need to be assessed.

In atmospheric chemistry, we were used to assimilate level 2 products of ozone (e.g. Massart et al., 2012; Emili et al., 2014; Peiro et al., 2018). Only recently, the direct assimilation of IASI radiances has been introduced in our chemistry transport model (Emili et al., 2019). Implementing a bias correction scheme requires careful diagnosis of the bias from observations monitoring. On the other hand, choosing an anchor demands also particular care and the choice depends on the full set of assimilated instruments. In this work, which is not based on a preexisting operational setup, we do not assimilate other ozone instruments than IASI. Thus, we had to assume that our observations are unbiased and we did not perform any bias correction. This assumption was adopted in many chemical analyses' studies before (e.g. Emili et al., 2019; Massart et al., 2012). Maintaining a similar framework allows a fairer comparison to these studies and might serve as a base for a future investigation of bias correction procedure for IASI.

We have modified the paper to include this discussion (L8 P7).

3.2. *were significant biases observed in the IASI observations?*

It is true that to properly answer this question, we need to compare our observations to a set of independent data. These data might be derived from other instruments or from models assumed to be unbiased. In our case, the absence of an independent instrument's data that are dense enough (spatially and temporally), on one hand, and the model that is biased on the other hand (see Figure 4 in Emili et al., 2019) make this approach (comparison with independent data) difficult to perform. To give a broad view of the bias in our system, we suggest here to show observation-background (O-B) statistics for six separate channels picked arbitrarily from the used band.

Figure 7_R2 shows the O-B statistics (averaged daily) as a percentage of the daily average of observations of each channel. We note that O-B varies between 0.6% and 1.7% over the observations for the six channels showed here. The same behavior of O-B statistics as percenatge of the background of each channel can be observed in Figure 8_R2. Thus, we can conclude that O-B is not too large compared to the background and observations values.

Nevertheless, the O-B statistics might not reflect a real bias of IASI observations since our model can biased. To address carefully this question and detect the bias of the IASI observations, an independent study is required. As we pointed above (comment 3.1), to maintain the same framework of the previous works (Emili et al., 2019) and aiming to evaluate only the contribution of the observation-error covariances, we have assumed that our observations are unbiased. Yet, the bias correction procedure of IASI observations in our case (atmospheric chemistry) should be investigated in future work.

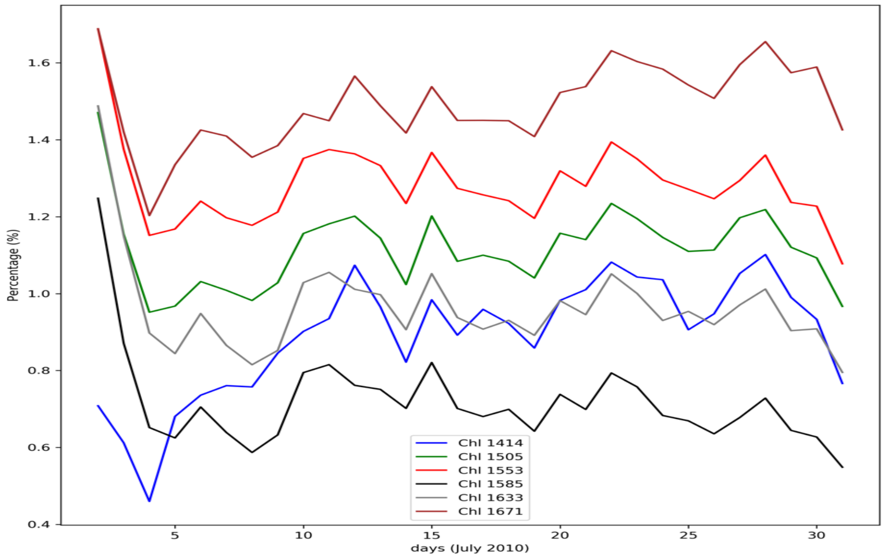

Figure 7_R2: The percentage of O-B statistics (averaged daily) over the daily average of observations of each channel (1414, 1505, 1553, 1585, 1633, 1671).

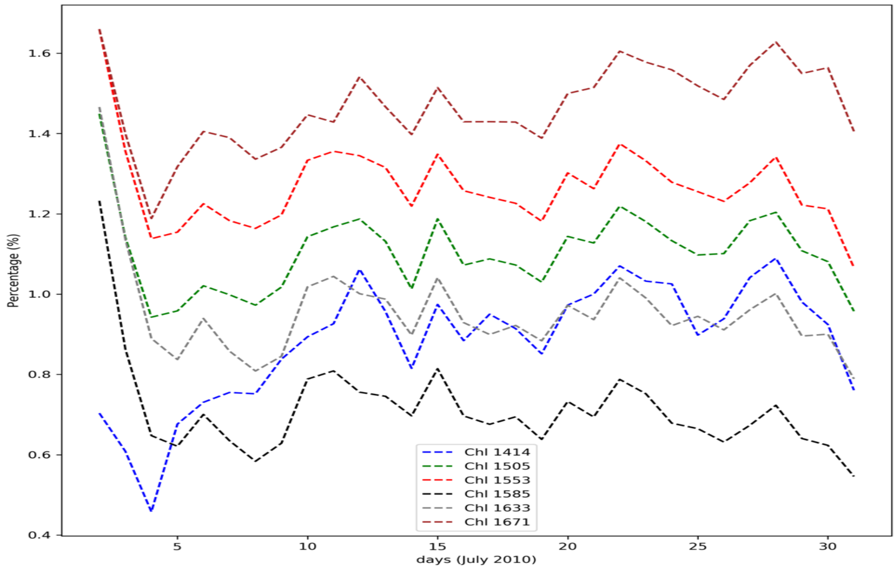

Figure 8_R2: The percentage of O-B statistics (averaged daily) over the daily average of the correspondent background of each channel (1414, 1505, 1553, 1585, 1633, 1671).

4. *The assimilation experiments were run for a 1-month period. Is this long enough to quantify the significance?*

It is certainly true that the longer the period of the study, the more significant the results. However, our main objective was to verify if an update of the observations error can have an impact in the ozone analysis accuracy, and our reference analysis is the one-month experiment already discussed in Emili et al. (2019). We show in the paper that the impact is significant in terms of ozone concentration. We also show that scores are globally improved against three set of independent validation observations (ozonesondes, MLS and OMI) with very different coverage and accuracy during both summer and winter (northern and southern hemisphere). The statistical significance of these results for the month of July 2010 is hence ensured. Nevertheless, extending the period of the experiment is important to verify the robustness of the approach and it is one of our perspective for the future. Indeed, Emili et al., 2020, have used a correlated matrix (as in the paper) to assess the impact of IASI measurements on global ozone reanalysis for a duration of one year (personal communication, manuscript already submitted to Geoscientific Model Development).

5. *Are SBUV available for assimilation or validation?*

Indeed, the SBUV and MLS might be assimilated for more accurate ozone analysis, at least in the stratosphere. However, we wanted to evaluate, through this study, the impact of accounting for interchannel error correlations of IASI in the assimilation system. Considering other accurate instruments might alleviate (or hide) the impact of IASI error covariance matrix on the analysis (as shown by Emili et al. 2019). For the validation we chose to validate with MLS rather than SBUV since it provides a continuous (during day and night) monitoring of the ozone as the infrared measurements with better vertical resolution.

II. Specific comments:

1. *P2 L4 and P5 L5, IASI is on Metop-A, B and C. But only Metop-A was available during the period of this study.*

This comment was included, we have specified that data from Metop-A were used. MetopB and C were not available in 2010 (the period of the study).

2. *P2 L16 Other references exist that discuss sources of IR error and error correlations. Representivity errors can also contribute to inter-channel error correlations.*

This part of the paper was modified to include some other references:

"Bormann et al. (2009) has listed …. of quality control in the data assimilation system" is replaced by:

"The interchannel error correlations might originate from observation operator errors. They can also arise from the instrument calibration and some practices of quality control (Bormann et al. (2009), Waller et al. (2016), Geer at al., (2019)). The representation errors (e.g. Janjić et al., 2018) may also introduce correlations."

3. *P2 L31 Is the main objective to study the impact on ozone analysis accuracy? Also, I think it is worth mentioning here that this is within the framework of a CTM.*

Yes, the main objective of our work is to improve the ozone analysis. We, therefore, modified the introduction (L31 P2 of the new version) to include this comment:

The sentence: 'The main objective of this study is, thus, to estimate the observation error covariances for IASI ozone-sensitive channels and to evaluate their impact on the analysis accuracy' is replaced by:

'The main objective of this study is, thus, to improve the ozone analysis accuracy within a chemistry transport model, by the main of using more realistic observation error covariances for IASI ozone-sensitive channels.

'their impact on the analysis accuracy' is replaced by 'their impact on the ozone assimilation within a chemistry transport model'

4. *P3 L1 There are numerous other studies that could be cited here.*

This line was modified to include some other references: (Weston et al. 2014, Bormann et al. 2016, Tabeart et al. 2020, Coopmann et al. 2020)

5. *P4 L22 Does the control vector include any other variables?*

No, it includes only SST and ozone. The paper was modified to precise that the control vector includes only SST and ozone.

This sentence: "The control vector includes the Skin Surface Temperature (SST) and the ozone." was replaced by:

"The control vector includes only the Skin Surface Temperature (SST) and the ozone."

6. *P5 L16. A brief discussion of channel selection is warranted. The abstract mentions that 280 channels are used, but this is worth restating here.*

We have added the band used:

"For this study, L1c data have been downloaded…" is replaced by

"For this study, a subset of 280 channels covering the spectral range between 980 and 1100 cm^{-1} was used. The channel selection is inherited from IASI Level 2 O3 retrievals (Dufour et al. 2011, Emili et al. 2019). L1c data have been downloaded…."

7. *P6 L21 What observations are being assimilated? Assimilating observations from OMI, SBUV or ozonesondes could help anchor the bias correction of IASI ozone channels.*

We have assimilated only IASI data. We have used OMI and ozonesondes to validate our results. Yes, a combination of ozonesondes and SBUV or OMI might serve as an anchor while processing the bias correction. However, we have assumed that our observations are unbiased as in many previous studies, and we have discussed this choice in bias correction comment (comment 3).

8. *P7 L18 Change "missing" to "absence." Is there a better justification for this assumption?*

This paragraph was entirely modified to include the bias correction discussion introduced in comment 3 (General comments).

9. *P8 L1 There are many other references that should be cited in addition to Stewart et al, 2009. In addition, there have been a few theoretical studies on the Desroziers method that could be cited here.*

This line was modified to include some other references: (Bormann et al. 2016, Waller et al., 2016, Tabeart et al. 2020, Coopmann et al. 2020)

10. *P8 L8 How many days of data were used in the computation? How many days of data were used for the re-estimations?*

We have used the same month (July) of the study for the re-estimation. In fact, the objective was to avoid to use an analysis that came from a diagonal R-matrix.

11. *P8 L9 The term "positive definite" is typically not used when discussing asymmetric matrices. A symmetric matrix is positive definite if and only if all of its eigenvalues are positive. I suggest that you instead discuss the eigenvalues after the matrix is made symmetric.*

Actually, we have made the estimated matrix symmetric by adding to it its transpose divided by 2: (0,5* $(R+R^T)$). Then we started to discuss the eigenvalues.

12. *P8 L14 Citing Van Loan seems out of place here. Is there a specific page number? This again is an incorrect citation, Gene Golub was another author of this textbook. I believe this method was used in Weston et al 2014 and in Tabeart et al, 2020b, so these might be better references.*

Yes, the referee is wright. We have inverted, by mistake, the citations here. Also, Van Loan is wrongly cited.

We have modified this part to include this comment.

This part:

Then we impose the negative eigenvalues to be equal to the smallest positive eigenvalue (Charles F. 15 Van Loan, 1996). Another method was tested here to recondition the estimated matrix, the one called *ridge regression* (Weston et al., 2013; Tabeart et al., 2020b) which consists of increasing all eigenvalues of R by the same amount. We favored the first method since the standard deviation and the correlation values remain closer to the initially estimated quantities.

Was replaced by (L11 P9 of the new version):

Then we impose the negative eigenvalues to be equal to the smallest positive eigenvalue as in Weston et al., 2013 and Tabeart et al., 2020b. Another method which consists of increasing all eigenvalues of R by the same amount was tested. We favored the first method since the standard deviation and the correlation values remain closer to the initially estimated quantities.

13. *P8 L22 Positive definite- was the re-estimated matrix symmetric?*

No, we have made it symmetric by adding to it its transpose divided by 2: (0,5* ($R+R^T$)).

14. *Figure 1- Please state in the caption what the "previous studies" are.*

Emili et al. 2019 was added to the Figure.

15. *Figure 2, 5- I do not trust the tick labels on the x and y axes. In Figure 1, there seems to be a gap in channels between 1010 and 1020 cm-1. If the ticks on Figure 2 are linear then the plotting program might be interpolating the covariances between 1010-1020 cm-1, or the tick labels could be wrong. I know that Matlab for example does not label ticks correctly by default when making matrix plots like these.*

Indeed, the tick labels were wrong. We modified this figure and we show the result below.

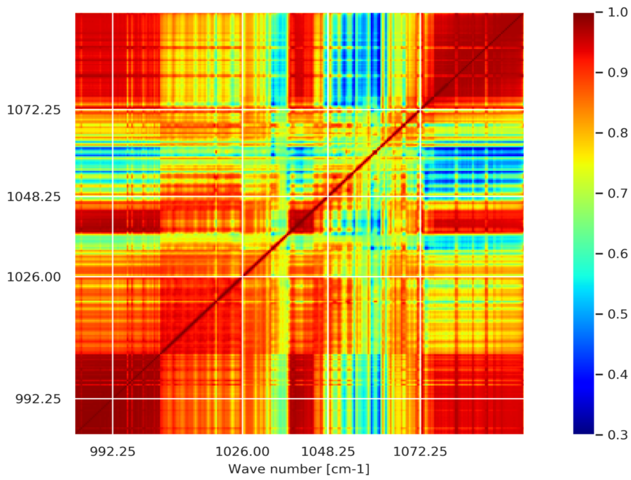

Figure 9_R2: R-matrix estimation over the globe (sea and land).

16. *in the caption, "statistics of Desroziers" would read better as "Desroziers method" or "Desroziers diagnostic"*

"statistics of Desroziers" is replaced by "Desroziers method"

17. *Figure 5- Anti-correlations likely exist over land. I suggest changing the colorbar scale to include negative values.*

In fact, since the ozone-sensitive and SST-sensitive channels present high interchannel correlations in this spectral window, we set the limits of the correlations between 0.3 and 1 to improve the information content of the figures. Also, no negative values were encountered in Figure 5.a, 5.b, and 5.c (of the old version of the paper). For Figure 5.e and Figure5.d (the differences in the old version of the paper) we took the absolute value of the differences divided by the global estimation.

We present below Figure 10_R2 (10b_R2, 10c_R2) the same Figure 5 (b and c) of the old version of the paper with -1.0 and 1.0 as limits. The same behavior was encountered in the Figure 5a (of the old version). Please note that the Figure 5 (of old version) was removed from this new version of the paper.

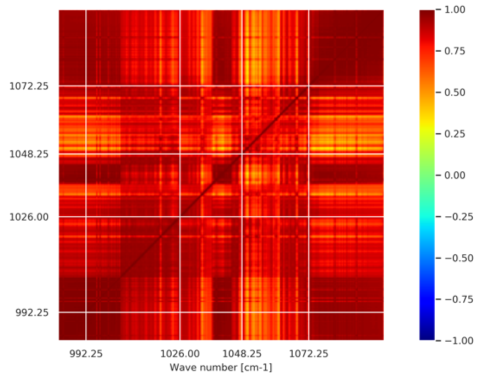

Figure 10b_R2: Estimation over the sea.

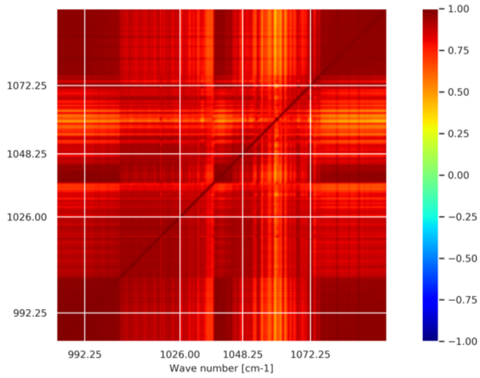

Figure 10c_R2: Estimation over the land.

18. *P13 L7 Suggest to specify in this sentence that this is an ozone analysis.*

… between the zonal average of the ozone analysis

19. *P13 L14 Why is the reduction important?*

In fact, by 'important' we wanted to say 'large'. We have replaced important by 'large'.

20. *P15 L10-14. I do not understand this discussion. What is the "estimation?" Why did the minimization fail to converge when using the "first estimation" but not the "second estimation?"*

Indeed, this discussion was not written in a way which allows a clear understanding. The first estimation did not fail to converge but needs more iterations than other estimations to converge.

We give, below, more details about what we meant by this discussion. We modified the paper to include this comment (L4 P14 to L13 P14).

We wanted to discern the imapct of the variance of that of the correlations on the convergence speed. To this end, we have performed three assimilation experiments using different R-matrices:

1st experiment: the employed R is estimated from the analysis computed using a diagonal R. The minimizer takes generally more than 100 iterations to converge.

2nd experiment: We use the analysis given by the 1st experiment to estimate another R-matrix (called second estimation in the old version of the paper). We have used this estimation to run another assimilation cycle. We have noticed that the minimizer needs about 60 iterations to converge.

3rd experiment: We have modified the R-matrix of the first experiment: we kept its correlations and replace its standard deviation with that of R used in the second experiment. We have noticed that the minimizer needs less than 100 iterations to converge (about 70 iterations).

Actually, using the first estimation of R, the minimization needs more than 100 iterations to converge, whereas about 60 iterations are needed with the second estimation of R. The results of the 3rd experiment seem to suggest that updating the variance has a larger impact on the convergence.

21. *Figures 9 and 10, in the captions it would be helpful to state that negative values indicate an improvement in fit relative to ControlExp. Is it possible to remove the empty space in Figure 10? What is meant by "divided by the average profile of radiosoundings?"*

21.1. Figures 9 and 10, in the captions it would be helpful to state that negative values indicate an improvement in fit relative to ControlExp. Is it possible to remove the empty space in Figure 10?

The figure was modified and included in the paper.

21.2. What is meant by "divided by the average profile of radiosoundings?"

We have replaced 'relative' by 'normalized' in the new version of the paper. We remind here how we computed RMSE presented in the figure 6 and 7 (of the new version of the paper).

- Figure 6: **(RMSE (control) − RMSE (exp))/radiosoundings**
- Figure 7: **(RMSE (control) − RMSE (exp))/MLS**

Where:
- 'exp': stand for RdiagExp (blue) and for RfullExp (green).
- 'radiosoundings': the average profile of the ozonesondes.
- 'MLS': the average of the MLS profiles.
- 'control': control experiment.

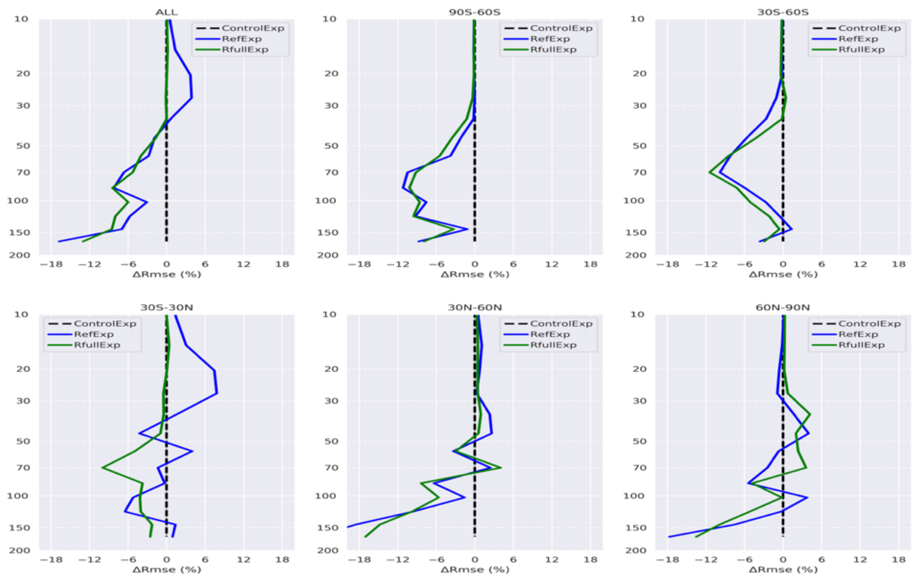

Fig 6_R2: Normalized difference of the RMSE with respect to the MLS for the R estimated (green) and R diagonal (blue). The normalized difference of the RMSE was computed by subtracting the RMSE of the analysis from the RMSE of the control for each experiment, divided by the average profile of the MLS. Negative values mean that the assimilation improved (decreased) the RMSE of the control simulation, and positive values indicate degradation (increase) of the RMSE

22. *P20 L16 Specify that ozone is added/reduced in the analysis.*

The word 'analysis' was added to specify that is the ozone analysis.

23. *P20 L17 "The total column also. . ." This sentence can be omitted. Isn't the total column result the validation against OMI mentioned in the next sentence?*

Yes, Indeed. The sentence is omitted.

III. Technical comments:

1. *The work from Desroziers is not cited correctly. It is from 2005, not 2006. Weston et al. is cited incorrectly as well; this paper is from 2014.*

   Corrected

2. *Throughout the article, ozone is written as O3, when it should be O3 with the 3 in the subscript.*

   Corrected

3. *P1 L22 Change "Remote sounding from satellites is" to "Remote soundings from satellites are"*

   Corrected

4. *P2 L1 Change "monitoring atmospheric gases, a large" to "monitoring of atmospheric gases. A large"*

   Corrected

5. *P2 L10 Should "Recent studies" be "A recent study" instead?*

   Corrected

6. *P2 L26 Remove (Weston et al 2013) from this sentence. All of the studies mentioned above show this result.*

   Corrected

7. *P2 L29 "(increase of the errors. . .)" The errors of what?*

   Corrected

8. *P3 L3 and other places. This should read "Desroziers method", not "Desroziers statistics"*

   Corrected

9. *P3 L7 Why not mention Section 5 in this paragraph?*

   Corrected
   'Then, the impact on data assimilation is reported in section 4 and validation against independent data is discussed in section 5'

10. *P4 L10 "aerosols" should be singular.*

Corrected

11. *P4 L18 What data file do you mean? I don't see one defined.*

Corrected

Indeed, we did not mean here by previously that it was cited in the paper. We meant by 'previously' that is given as an input to the experiment.

We have modified this sentence.

12. *P4 L23 "evaluate the impact of the estimated observation error" and the error covariances, correct?*

Corrected
'evaluate the impact of the estimated observation error covariances on the analysis'

13. *P4 L26 Change "reminded" to "given"*

Corrected

14. *P6 L13 Change this sentence to "...carried by a radiosonde continuously transmitting measurements as it ascends."*

Corrected

15. *P6 L26 Change "found out" to "found"*

Corrected

16. *P7 L26 Change "radiative transfer may" to "radiative transfer model may" and "statistics of error from the instruments" to "error statistics from instrument"*

Corrected

17. *P7 L29 The second term in the expected value should be a vector transpose.*

Corrected

18. *P8 L7 "(with a standard. . ." there is no closing parenthesis*

Corrected

19. *P8 L12 Change "assumed" to "applied"*

Corrected

20. *P8 L15 Change "An other" to "Another"*

Corrected

21. *P8 L 20 Change "The resulted standard deviation was greater than the one" to "The resulting standard deviations were greater than those"*

Corrected

22. *P9 L 22 Figure 3 shows standard deviations, not differences.*

Corrected
This figure was omitted in this version of paper.

23. *P14 L1-2 These two sentences are unnecessary*

Corrected: sentence omitted

P15 L25 This sentence should be a part of the previous paragraph.

Corrected

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