Answer to the referee 1

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 it.

The answer to the comments is organized as follows. First, we list some notations that will be adopted in the answers and the major 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 '**_R1'** added in the legends of the figures: means referee 1.

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 3).
- We have modified Figure 7 (of the old version) to show data averaged by grid box as suggested by the referee.
- 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. *A guideline is missing. What is the main objective of this paper? To diagnose an R-matrix or to improve ozone analyses using a diagnosed R-matrix?*

   Answer

   It is true that the way we have defined the main objective of this paper in the introduction (L31 P2 of the new version) might cause confusion between both estimating R-matrix or improving ozone analyses. As a matter of fact, the main objective is to improve the ozone analyses, by the mean of using more realistic observation error covariances. Estimating and discussing the R-matrix was not an end in itself, it is used to improve the assimilation of IASI radiances.

   In this new version, we have reduced the discussion of the diagnostic results (land/sea & day/night) to put much more emphasis on the main objective: ozone analyses.

   We modified the paper to include this comment while defining the objective (L31 P2 of the new version).

2. *The paper lacks a discussion about the bias correction that may be needed for ozone-sensitive channels. A comparison with the work of (Han and McNally, 2010) would have been relevant:*

   Answer

   We agree on the fact that the discussion of the bias correction was not well detailed in the paper. We give here more details, and we modified the paper to include this discussion (L8 P7).

   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. *The comparisons between observation- errors according to surface types and between day and night are interesting:*

Here we want to remind the referee about a change that we have introduced to the new version of the paper following one of the referee2's comments (referee 2, 1st comment):

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 by the referee 2, we cut the figures of day/night and sea/land (Figure 3, 4 and 5 of the old version). We gave more details about this choice in the answer to the comment 1 of the referee 2 answers.

We address the referee to check the referee2's answer for more details.

4. *I wonder about the significance of an experiment of one month. It would have been beneficial to continue these experiments over 2 months, as well as over two distinct periods (summer and winter)?*

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).

II. Specific comments:

1. *Specify in the title, that this work is carried out in a chemistry transport model.*

Answer:

New title: 'Estimation of the error covariance matrix for IASI radiances and its impact on the assimilation of ozone in a chemistry transport model.'

2. *P1, L6: (...between 980 and 1100 cm-1) I suggest adding that this spectral range includes ozone-sensitive channels and atmospheric window channels.*

Answer:

We used a subset of 280 channels…to estimate the observation error covariance matrix. This spectral range includes ozone-sensitive channels and atmospheric window channels. We computed hourly …

3. *P1, L11: (The computational cost...) This sentence is useless without explanation. I suggest you delete it or add a short comment.*

Answer:

The computational cost was …in the assimilation system, by reducing the number of iterations needed for the minimizer to converge.

4. *P2, L30: (. . .impact on analysis accuracy.) Specify that this is the impact on the ozone analysis.*

Answer:

impact on the ozone analysis accuracy.

5. *P3, L2 There are more recent studies on the same subject that you can reference: (Weston et al. 2014, Bormann et al. 2016, Tabeart et al. 2020, Coopmann et al. 2020)*

Answer

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)

6. *P3, L29: (. . .the radiative transfer model RTTOV) Most recent reference to the work of (Saunders et al. 2018).*

Answer

Saunders et al. 2018 was added in the references.

7. *P3, L31: (...Starting from an atmospheric...) Specify that RTTOV requires a vertical temperature and humidity profile.*

Answer

Replace "Starting from an atmospheric vertical profile" by "Giving an atmospheric profile of temperature, water vapour and, optionally, trace gases, aerosols and hydrometeors, together with surface parameters and a viewing geometry", RTTOV simulates….

8. *P4, L6: What about other chemical variables (CO2, CH4, CO, N2O, SO2)? Do you use reference profiles? Which coefficient file do you take into account?*

Answer

These chemical variables (CO_2, CH_4, CO, N_2O) were set to the reference profiles of RTTOV. For the coefficient file, we used the coefficients for v9 predictors computed on 101 levels.
The SO_2 was not available in RTTOV v11 (used in this study).
We have added this comment to the paper (L13 P4 of the new version)

9. *P3, L17: Indeed, the observation-error variances and observation-error covariances plays a fundamental role in the data assimilation process. In addition, background- errors are also very important in this process. For the purpose of consistency, it is required, at least, to show the background-error variances or background-error error standard deviation, as well as, the background-error correlations matrix.*

Answer:

We have plotted the background-error standard deviation in % of the background profile (Figure 1_R1) and the zonal error correlation length scale Lx (Figure2_R1).

In the paper, we prefer not to show these figures since they are not very informative but we have added the background-error description in the table 1 of the new version of the paper.

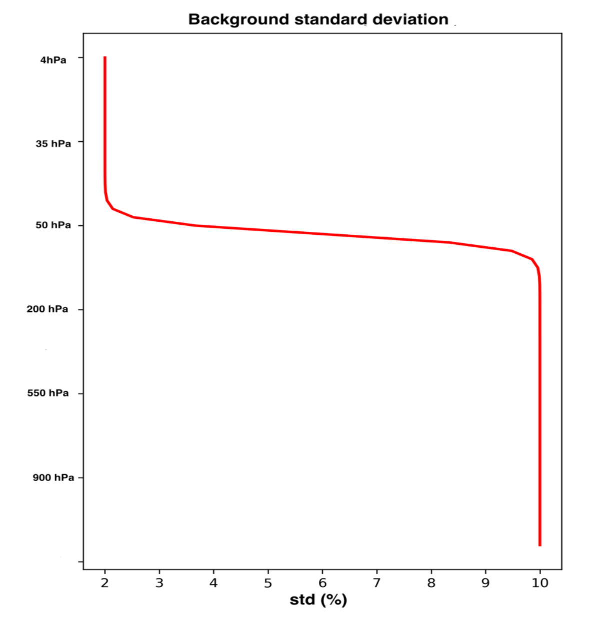

Figure 1_R1 : background-error standard deviation (square root of the diagonal of B) in % of the background profile.

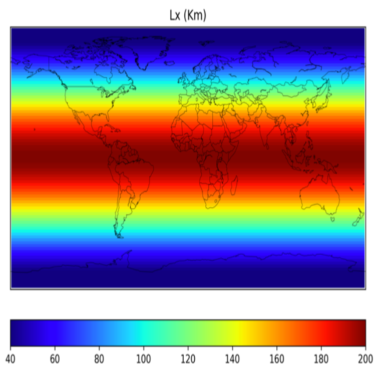

Figure 2_R1: The zonal error correlation length scale (Lx).

10. P4, L19: (as a percentage of the observation values.) What does this percentage look like?

Answer:

The referee is right, this sentence was not very clear. In the beginning of this paragraph, we wanted to list first all the possibilities offered by MOCAGE.

Since in our study we define our observation's error covariances in a file (as input), we omit this sentence and we keep only the case we are using (R-matrix read from a file).

This paragraph was omitted.

'In the data assimilation system of MOCAGE, the observation error covariance matrix can be read from the data file previously defined. In the case of diagonal matrix, the variances can be calculated as a percentage of the observation values.'

11. P4, L22: Are there other variables included in the control vector?

Answer:

No. The control vector contains only ozone and surface skin temperature. The word 'only' was added to this sentence on the paper.

12. P5, Table 1: Can you provide more information about the ozone background?

Answer:

This column was added to table 1.

Ozone background	Hourly 3D forecasts of MOCAGE.

13. P5, L15: (. . .co-located land mask...) Wouldn't it be the "Land Sea Mask" instead?

Answer:

Yes, this line was modified '…Data files also contain the co-located land sea mask and cloud fraction values…'

14. P5, L16: In this case, from which satellite platform are IASI observations extracted? MetopA, B, C?

Answer

Data are extracted from the MetopA platform. MetopB and C were not available for the period of the study.

15. P6, L20: Another reference to (Emili et al. 2019) It would be very useful to summarize the configuration of the experiments in a table.

Answer

In this new version of the paper, we extend Table 1 to include other elements of the experiment's configuration.

Below, we present lines added to the Table 1 of the paper.

Period of the study	July 2010
Background error	Vertically variable and computed as % of the background profile (using a value of 2% above 50 hPa and 10 % below).
Background error zonal correlation	Exponential with a length scale set to 200 Km and reduced towards the pole to account for the

	increasing zonal resolution of the regular latitude-longitude grid.
Background meridional error correlation	Exponential with a length scale set to 200 Km.
Background error vertical correlation	Exponential with a length scale set to 1 grid point (vertical level).

16. *P6, L27: Can you compare these ozone background-error standard deviation with other values used in recent research?*

Answer:

The ozone background-error standard deviation was taken as percentages of the background O_3 profile. This strategy was adopted previously by many studies (e.g. Emili et al., 2014, Peiro et al., 2018, and Emili et al., 2019). Emili et al., 2014 and Peiro et al., 2018 have used a percentage of 15% in the troposphere and 5% in the stratosphere.

In this study, we have adopted a detailed chemical scheme (chemical scheme combining both Regional Atmospheric Chemistry Mechanism for the troposphere (Stockwell et al., 1997) and REPROBUS (Lefevre et al., 1994) for the stratosphere). This scheme was shown to reduce the model bias compared to scheme used in Emili et al., 2014 and Peiro et al., 2018 (see Figure 4 in Emili et., 2019). Hence, we chose the same background errors as in Emili et., 2019: 2% of the O_3 profile above 50hPa and 10% below. An important reason to keep the background errors similar to the setup of Emili et al. (2019) is also that we wanted to examine here exclusively the impact of **R**, as already reminded in the introduction and in the conclusion.

The paper was modified to add this discussion (L16 to L22 P6).

This part of the paper (P6 L27 to L29 of the old version):

"The background standard deviation was, thus, taken equal to 2% above 50 hPa and 10 % below to mimic the validation's behavior. Similar choices were employed in (Massart et al., 2012; Peiro et al., 2018).

Was replaced by (L16 P29 of the new version) by:

"This strategy was adopted previously by many studies (e.g. Emili et al., 2014, Peiro et al., 2018, and Emili et al., 2019). Emili et al., 2014 and Peiro et al., 2018 have used a percentage of 15% in the troposphere and 5% in the stratosphere.

In this study, we have adopted a detailed chemical scheme. This scheme was shown to reduce the model bias compared to scheme used in Emili et al., 2014 and Peiro et al., 2018 (see Figure 4 in Emili et., 2019). Hence, we chose the same background errors as in Emili et., 2019: 2% of the O_3 profile above

50hPa and 10% below. An important reason to keep the background errors similar to the setup of Emili et al. (2019) is also that we wanted to examine here exclusively the impact of R, as already reminded in the introduction and in the conclusion."

17. P7, L11: On what criteria were these channels identified as sensitive to water vapor?

    The channels sensitive to water vapor in the ozone band have been identified by previous studies on IASI trace gases retrieval using RT simulations (Barret et al 2011, see also Fig. 1 of https://acp.copernicus.org/articles/11/857/2011/acp-11-857-2011.pdf). We used here the same channel selection of previous O3 studies.

18. *P8, L18 to L28: This paragraph is complicated to follow and it is a pity because it is important for the next step. I suggest you summarize the different configurations in a table.*

We have redrafted this paragraph (L15 to L24 P9 of the new version of paper) to improve the clarity of the discussion of different estimations and the one used in the paper. This paragraph was replaced by:

"Using outputs (analyses and forecasts) derived from 3D-Var experiment that uses a diagonal R-matrix (called hereafter 1st 3D-Var experiment) in the estimation process might have an impact on the diagnosed R-matrix. The matrix derived using these outputs is called hereafter 1st estimation. We performed another 3D-Var experiment (2nd 3D-Var experiment) using the 1st estimation. The outputs (analyses and forecasts) of this experiment (2nd 3D-Var experiment) were used to estimate another R-matrix called 2nd estimation. The standard deviation of the 2nd estimation is larger than that of the 1st estimation (not shown). The same goes for correlations (not shown). It should be noted that the 2nd estimation was positive definite, unlike the 1st estimation where some unrealistic features were encountered. We have followed the same process to reestimate two other matrices (3rd and 4th estimation). The differences of the estimations in terms of standard deviation and correlations became smaller as we reestimate the matrices, suggesting a sort of convergence of the estimation.

We have adopted the 2nd estimation for the results shown in this work. The reason for this choice will be discussed later (section 5.2)."

We show below a figure summarizing this discussion. This figure was not added to the paper.

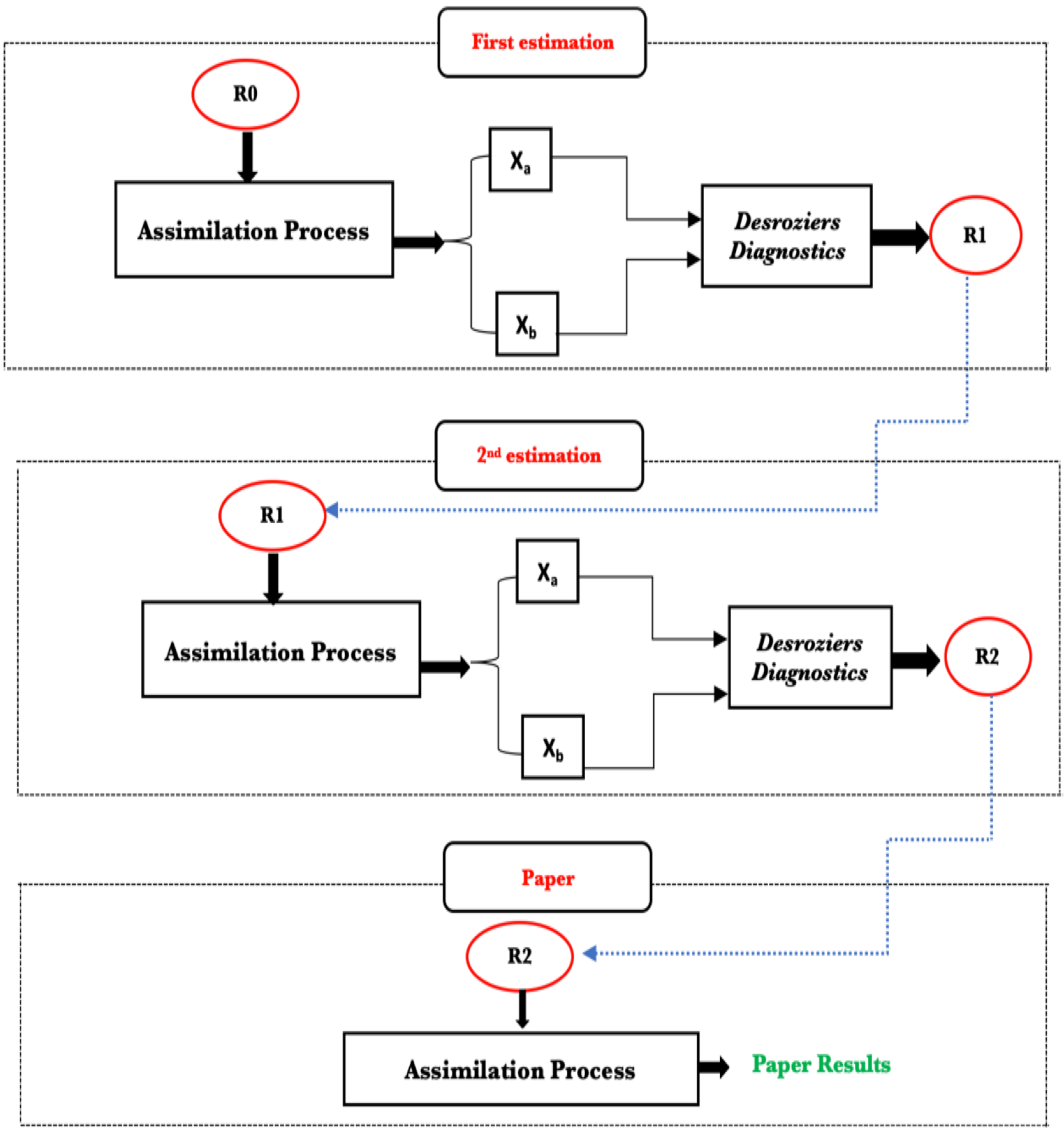

Figure 3_R1 : different estimations discussed in the paper and the one used for the results.

19. *P10, Figure 2: Correlation matrices can vary between -1.0 and 1.0. I expected to see negative correlations between some channels in the atmospheric window and some ozone-sensitive channels. Why not represent the matrix between -1.0 and 1.0, centered on white at zero?*

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. We present below Figure 4_R1 (the same as Figure 2 of the old version of the paper) with -1.0 and 1.0 as limits:

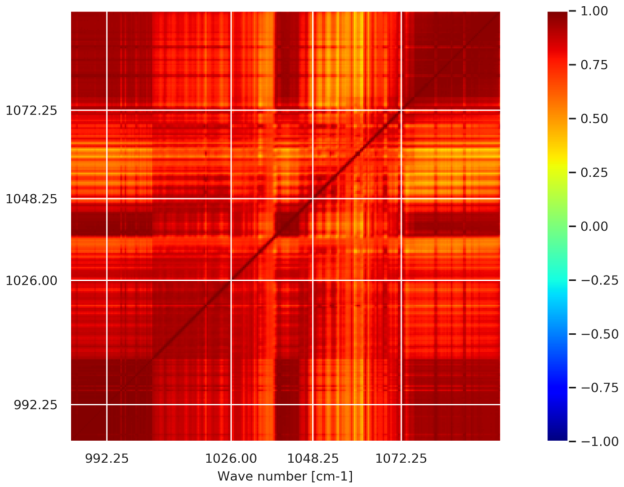

Figure 4_R2: Correlation matrix estimated over the globe (sea and land).

20. P12, Figure 5: Same remark as above about the color scale.

No negative correlations have been encountered in Figure 5.a, 5.b, and 5.c (of the old version of the paper). In fact, since the ozone-sensitive and SST-sensitive channels present high interchannel correlations, we set the limits of the correlations between 0.3 and 1. 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 show below Figure 5_R1 (5b_R1, 5c_R1) the same Figure 5 (b and c) of the old version of the paper with -1.0 and 1.0 as limits (Figure 5 a of the old paper is the same in the previous comment 19). Please note that the Figure 5 (of old version) was removed from this new version of the paper.

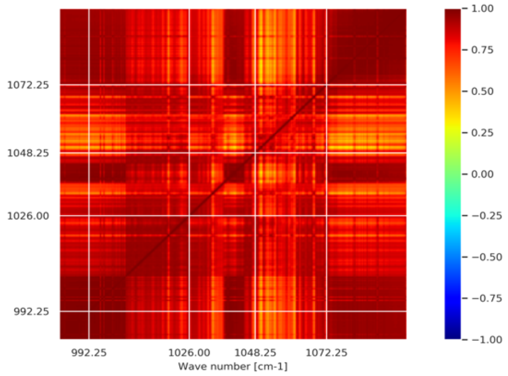

Figure 5b_R1: Correlation matrix estimated over the sea.

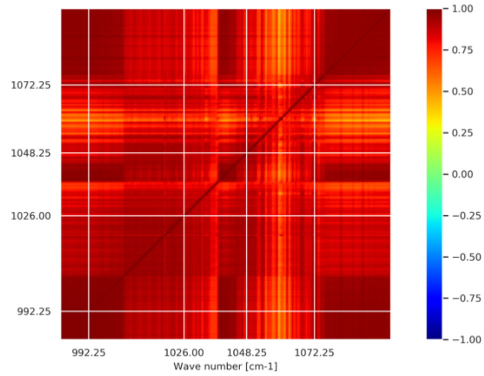

Figure 5c_R1 : Correlation matrix estimated over the land.

21. *P12, L8: The naming of the experiments is not appropriate because one could confuse Control and Reference. I would suggest RdiagExp instead of RefExp.*

Answer:

RefExp is changed to RdiagExp

22. *P14, L4: It would be useful to explain the physical link between skin temperature and ozone in the assimilation of infrared observations. Is there any consideration of inter- variable background-error correlations between O3 and Tskin?*

Answer:

Yes, indeed. The skin temperature is physically linked to the ozone measured. In fact, the skin temperature interacts with the ambient atmosphere. An increase of SST can for example create a convective movement impacting the transport of the ozone. However, the skin temperature is given only at the observation location in this study and it is specified with values interpolated from NWP forecasts (IFS), whereas ozone is a 3D field issued from the chemistry transport model. Hence, the estimation and potential account of error correlations between the two variables seems challenging in our system. We think that Earth System models where both skin temperature and ozone are modeled (and assimilated) might represent a preferable framework for analyzing this particular aspect.

In this work, we did not consider the background-error correlation that might exist between O3 and SST.
We have modified the paper to include this paragraph (L6 P12 of the new version of the paper).

23. *P14, Figure 7: There is also increase in difference on land using RfullExp, mainly in Africa and South America. This can be related to the differences in observation-errors depending on the surface. . . In addition, there are too many pixels on the map. It would be interesting to average by box in order to better exploit the information provided by this Figure.*

23.1. *There is also increase in difference on land using RfullExp, mainly in Africa and South America:*

Answer:

Indeed, the increase in difference over the land seems related to the dependence of observation-errors on the surface. In fact, the number of observations over the sea represents almost 70% of the total observations we have used in this study. Consequently, our SST analysis stays closer to background values (IFS forecasts) over the sea than over the land.
This comment was added to the paper (L4 P13).

23.2. *. It would be interesting to average by box in order to better exploit the information provided by this Figure.*

Answer:

Figure 7 (of the old version) is replaced by other figures below where we have averaged the observations by grid box.

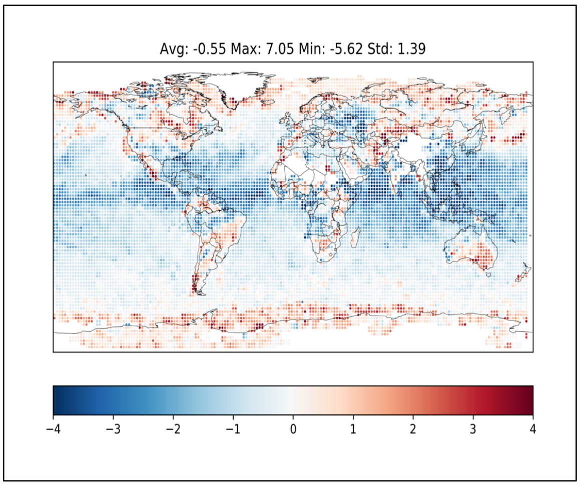

Fig7a_R1. Difference (in °C) between the IFS SST forecast and the analysis of the SST given by RdiagExp (averaged by grid box).

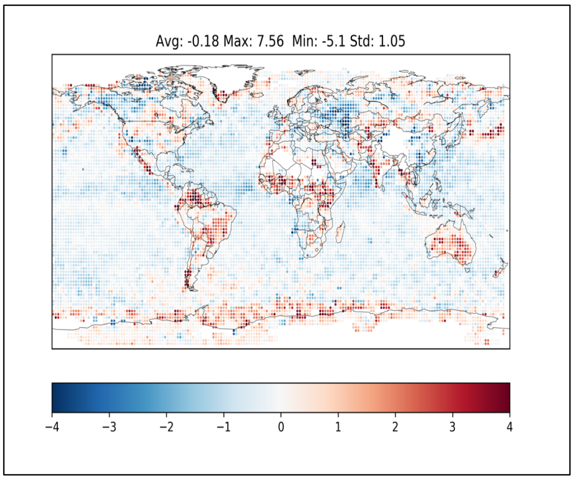

Fig7b_R1. Difference (in °C) between the IFS SST forecast and the analysis of the SST given by RfullExp (average by grid box).

24. *IASI channels between 1000 and 1070 cm-1 are mainly sensitive to ozone above 100 hPa, which poses the challenge of using other observations for a complete analysis of ozone over the entire atmospheric column...*

Yes, indeed. It would have been advantageous to assimilate other instruments (MLS for example in the stratosphere and ozonesondes for the free troposphere) for a complete analysis of ozone. However, we wanted to evaluate, through this study, the impact of accounting for interchannel error correlations of IASI in the assimilation system. Assimilating other accurate instruments might alleviate (or hide) the impact of interchannel observation-error correlations of IASI on the analysis, as it was shown in Emili et., al (2019).

25. *the high sensitivity of the ozone channels raises the problem of the amount of information remaining after a cloud detection...*

    Yes, it would be challenging to take into account the pixels affected by clouds (and including the corresponding cloud properties in the radiative transfer) during the assimilation of IASI channels for ozone. This might be an area of research for future work.

    The idea for this study was to keep the same configuration of the assimilation system adopted in the study of Emili et al., 2019, to be able to evaluate only the impact of an updated observation error covariance matrix in clear-sky conditions.

III. <u>Technical comments:</u>

1. *P1, L4: (Modèle de Chimie Atmosphérique à Grande Echelle)*

   Corrected

2. *Throughout the paper: I suggest (Chemistry Transport Model) instead of (Chemical transport model)*

P1, L5: (. . .already adopted in numerical weather prediction centers) This is not the case for all centers, (. . .already adopted in some numerical weather prediction centers)

   Corrected

3. *Throughout the paper: Beware of the systematic use of (Furthermore). Vary the ad- verbs.*

   Corrected

4. *P2, L8: (. . .to construct a realistic picture of the...) The term (picture) is not appropriate, I suggest changing the word.*

   Corrected

5. *P2, L22: (...uncorrelated, some Numerical Weather Prediction...) P2, L31: (. . .evaluate their impact on the ozone analysis accuracy) P3, L19: (. . .MOCAGE is fed with forced by meteorological...)*

   Corrected

6. *P5, L5: (. . .the polar-orbiting satellite Metop-A, B and C launched...)*

   Corrected

7. *P6, L24: (The ozone forecast-error standard deviation...)*

   Corrected

8. *P6, L27: (The ozone background-error standard deviation...)*

   Corrected

9. *P7, L1: (The ozone background-error covariance matrix. . .)*

   Corrected

10. *P9, L12: (. . .we present the diagnosed correlation matrix...)*

    Corrected

11. *Throughout the paper: Be careful to capitalize the words (Figure)*

    Corrected

12. *Throughout the paper: Write rather with dashes (observation-errors, background- errors , ozone-sensitive,...)*

    Corrected

13. *P15, L22: (. . .encountered in these regions in the stratosphere...)*

    Corrected

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Stockwell et al., 1997] Stockwell, W. R., Kirchner, F., Kuhn, M., and Seefeld, S. (1997). A new mechanism for regional atmospheric chemistry modeling. *Journal of Geophysical Research*, 102:25847–25879.

Tabeart, J. M., Dance, S. L., Lawless, A. S., Migliorini, S., Nichols, N. K., Smith, F., and Waller, J. A.: The impact of using reconditioned correlated observation-error covariance matrices in the Met Office 1D-Var system, Quarterly Journal of the Royal Meteorological Society, pp. 1–22, https://doi.org/10.1002/qj.3741, 2020a.

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.

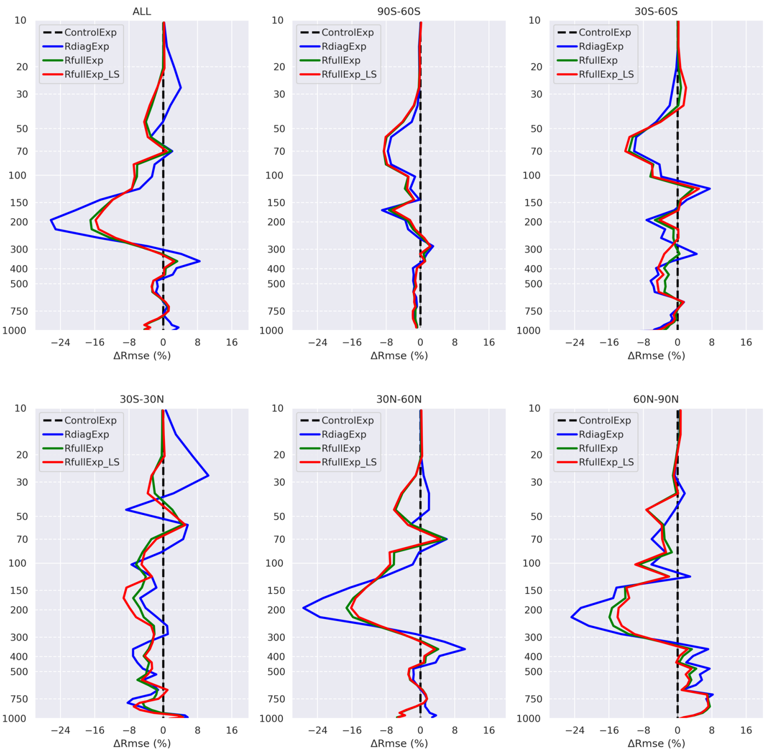

Figure 1_R2: Normalized difference of the RMSE with respect to radiosoundings for the RFullExp (green), RdiagExp (blue) and the RfullExp_LS (the experiment using separated matrix according to the type of the surface) in red. The difference of the RMSE was computed by subtracting the RMSE of the controlExp 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).

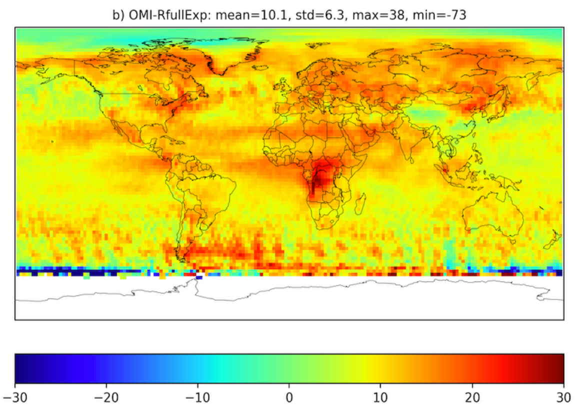

Figure 2_R2: Difference of the ozone total column (DU) provided by OMI and that of the assimilation experiment using a matrix estimated over the glob averaged over the month of the study.

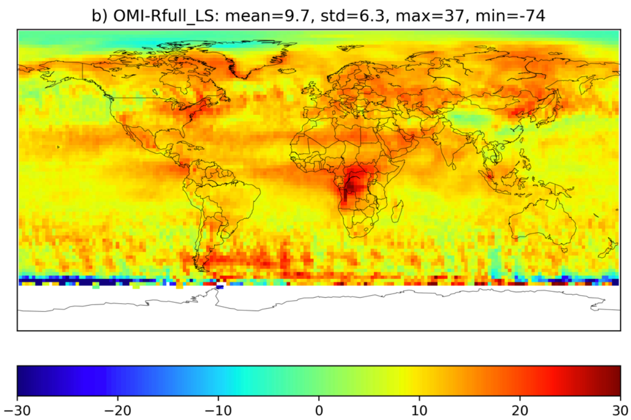

Figure 3_R2: Difference of the ozone total column (DU) provided by OMI and that of the assimilation experiment using a matrix according to the type of the surface (land/sea) averaged over the month of the study.

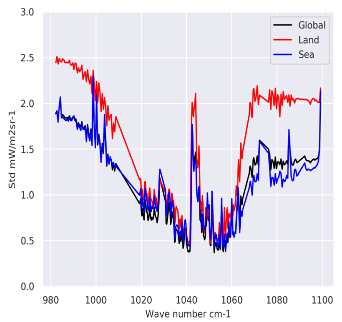

Figure 4_R2: Standard deviation estimated using Desroziers diagnostics according to the type of the surface (sea, land and global).

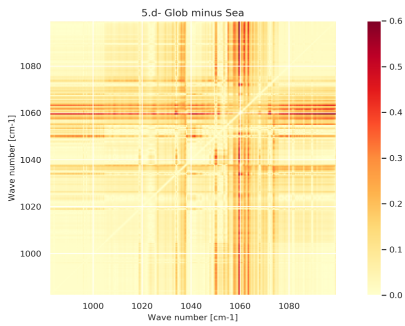

Figure 5_R2: Difference (in %) between global and sea correlation matrix (divided by the global matrix).

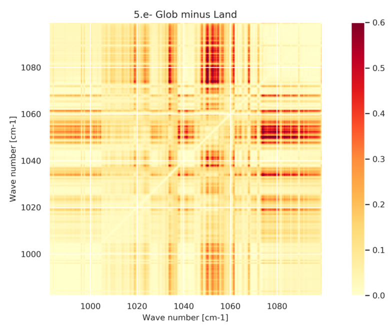

Figure 6_R2: Difference (in %) between global and land correlation matrix (divided by the global matrix).

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.

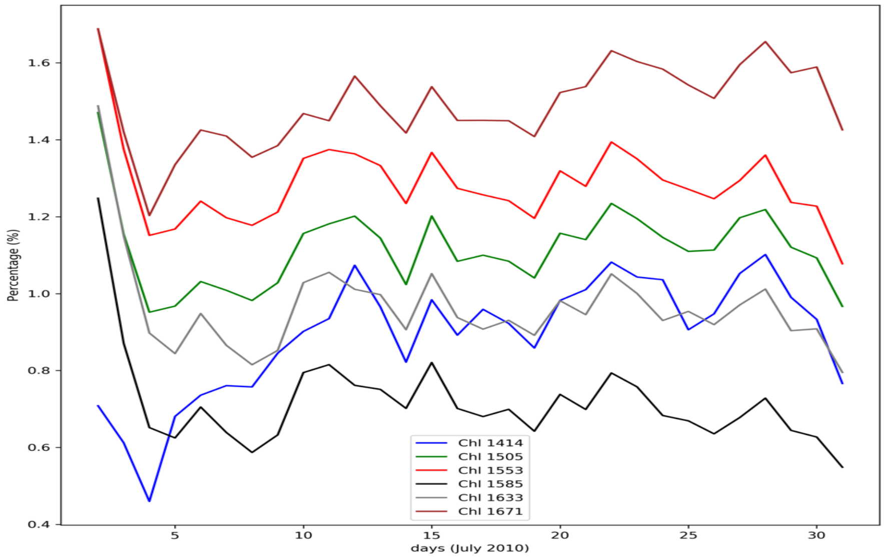

Figure 7_R2: The percentage of O-B statistics (averaged daily) over the daily average of observations of each channel (1414, 1505, 1553, 1585, 1633, 1671).

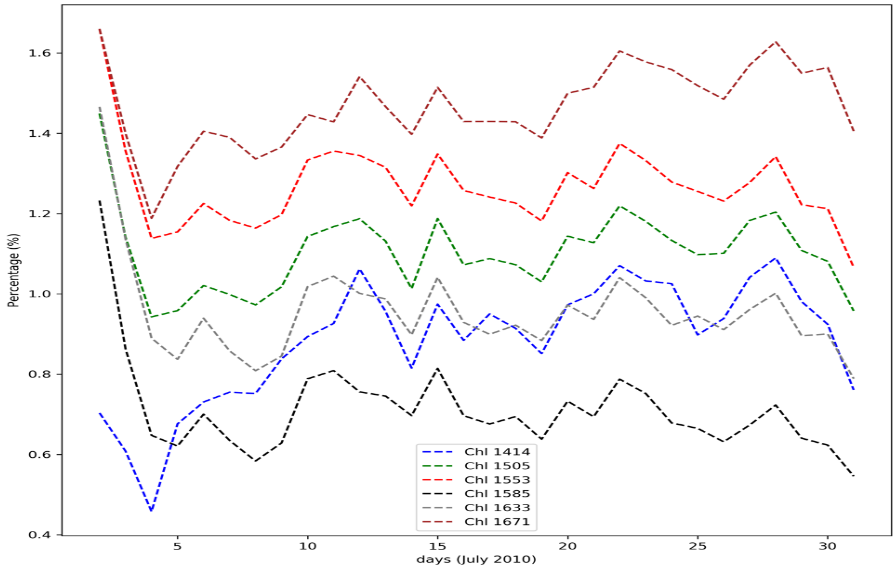

Figure 8_R2: The percentage of O-B statistics (averaged daily) over the daily average of the correspondent background of each channel (1414, 1505, 1553, 1585, 1633, 1671).

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. <u>Specific comments:</u>

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.

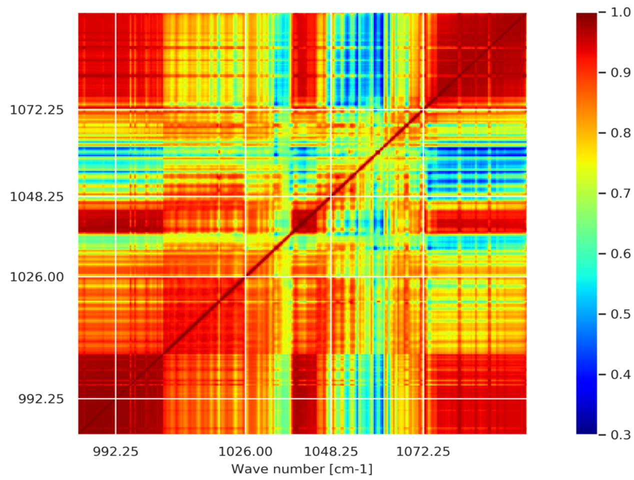

Figure 9_R2: R-matrix estimation over the globe (sea and land).

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.

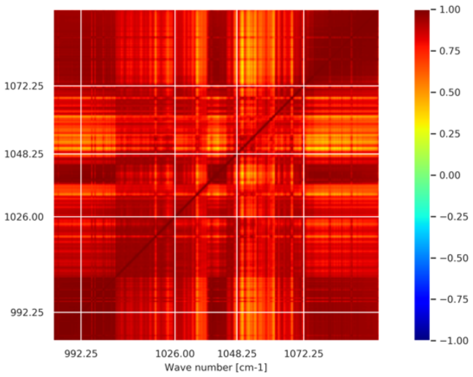

Figure 10b_R2: Estimation over the sea.

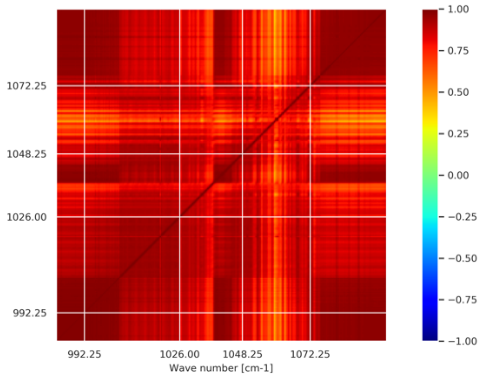

Figure 10c_R2: Estimation over the land.

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.

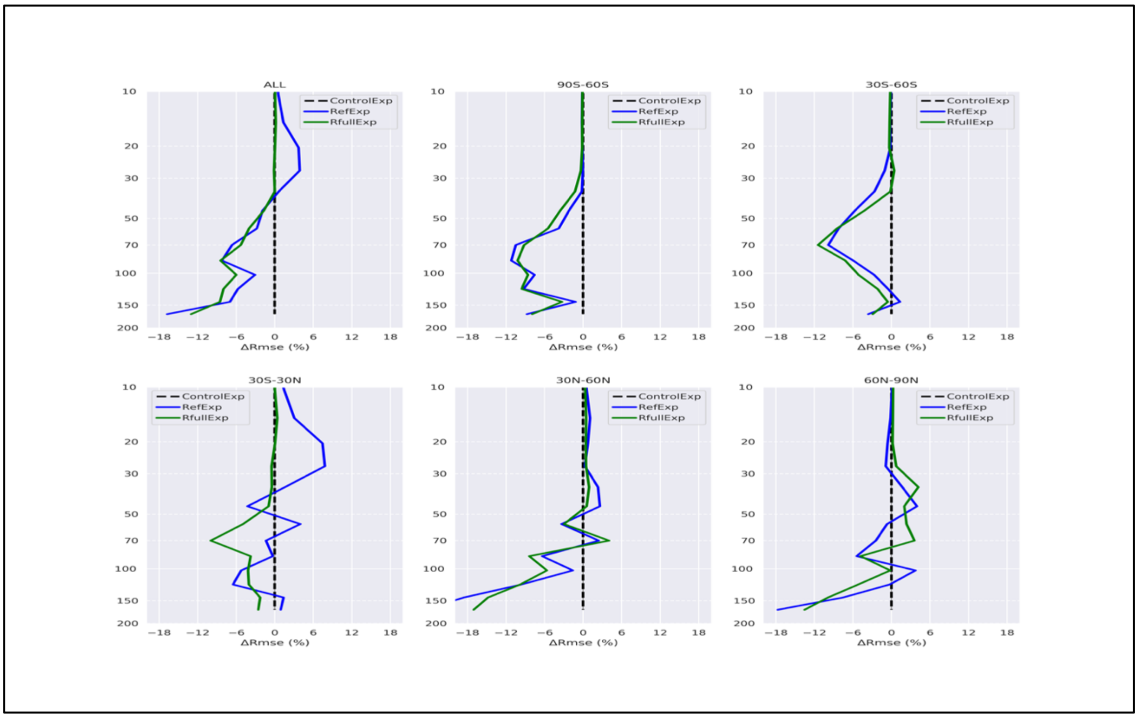

Fig 6_R2: Normalized difference of the RMSE with respect to the MLS for the R estimated (green) and R diagonal (blue). The normalized difference of the RMSE was computed by subtracting the RMSE of the analysis from the RMSE of the control for each experiment, divided by the average profile of the MLS. 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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Estimation of the error covariance matrix for IASI radiances and its impact on the assimilation of ozone [..*]in a chemistry transport model

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Abstract. In atmospheric chemistry retrievals and data assimilation systems, observation errors associated with satellite radiances are chosen empirically and generally treated as uncorrelated. In this work, we estimate inter-channel error covariances for the Infrared Atmospheric Sounding Interferometer (IASI) and evaluate their impact on ozone assimilation with the [..2]chemistry transport model MOCAGE (MOdèle de [..3]Chimie Atmosphérique à Grande Echelle). The method used to calculate observation errors is a diagnostic based on the observation and analysis residual statistics already adopted in [..4]some Numerical Weather Prediction centers. We used a subset of 280 channels covering the spectral range between 980 and 1100 cm^{-1} to estimate the [..5]observation-error covariance matrix. This spectral range includes ozone-sensitive and atmospheric window channels. We computed hourly 3D-Var analyses and compared the resulting O_3 fields against ozonesondes and the measurements provided by the Microwave Limb Sounder (MLS) and by the Ozone Monitoring Instrument (OMI).

The results show significant differences between using the estimated error covariance matrix with respect to the empirical diagonal matrix employed in previous studies. The validation of the analyses against independent data reports a significant improvement, especially in the tropical stratosphere. The computational cost has also been reduced when the estimated covariance matrix is employed in the assimilation system, by reducing the number of iterations needed for the minimizer to converge.

1 Introduction

Ozone is an important trace gas that plays a key role in the Earth's radiative budget (Iglesias-Suarez et al., 2018), in the chemical processes occurring in the atmosphere, and in the climate change (United Nations Environment Programme [UNEP] 2015). Tropospheric ozone also behaves as a pollutant with negative effects on vegetation and human health (UNEP2015, 2015). The stratospheric ozone is, nevertheless, a vital component of life on the Earth since it protects the biosphere from

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harmful ultraviolet solar radiation (WMO, 2014). Therefore, monitoring the atmospheric ozone has been a subject of numerous research studies and projects (e.g. Monitoring Atmospheric Composition and Climate (MACC) project (Inness et al., 2013)). O_3 surveillance is carried out through numerical forecast models and observational systems. The information arising from these two sources is, thereafter, combined with the data assimilation techniques to improve the system state and forecasts.

Remote [..[6]]soundings from satellites are an essential component of the observation's network (Clerbaux et al., 2009). Several remote sensors relying on thermal emission of the Earth and the atmosphere have demonstrated their ability to provide appropriate information for total columns or vertical profiles of atmospheric gases such as water [..[7]]vapour, carbon dioxide, and ozone [..[8]](Clarisse et al., 2008; Clerbaux et al., 2009; Irion et al., 2018). Furthermore, the role of thermal infrared sounders does not typically end at the monitoring [..[9]]of atmospheric gases. A large number of applications have taken advantage of these measurements: meteorological parameters (clouds, temperature, and humidity), climate change (e.g., [..[10]]MacKenzie et al. (2012)). Infrared Atmospheric Sounding Interferometer (IASI) is one of these thermal infrared sounders onboard [..[11]]MetopA which provides global scale observations for a series of key atmospheric species (Clerbaux et al., 2009).

Data assimilation has been introduced relatively recently in atmospheric chemistry, in the stratosphere layer (Fisher and Lary, 1995) and for the troposphere [..[12]](Elbern et al., 1997). Chemical fields estimated by [..[13]]chemistry transport models (CTM) are combined with observations to construct [..[14]]more accurate description of the atmospheric composition evolution (Lahoz et al., 2007). Numerous studies have been conducted assimilating satellite retrievals of ozone (Emili et al., 2014; Massart et al., 2009). However, the quality of analyses might be influenced by the prior information used for the retrievals. [..[15]]A recent study (Emili et al., 2019) attempted to assimilate satellite radiances directly in a [..[16]]chemistry transport model (CTM) to overcome this issue. In chemical assimilation systems that assimilate radiances directly, but also in most of the current satellite retrieval algorithms (Dufour et al., 2012), the observation errors are empirically adapted from the nominal instrumental noise and assumed to be uncorrelated. This assumption is questionable since we use a radiative transfer model that may introduce similar errors among different spectral channels [..[17]](Bormann et al., 2010). In other words, an error dependency between channels of the band used is likely to be introduced. [..[18]]The interchannel error correlations might originate from observation operator errors. They can also arise from the instrument calibration and some practices of quality control [..[19]](Bormann et al., 2010; Waller et al., 2016; Geer, 2019). The representation errors (Janjić et al., 2018) may

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[15]removed: Recent studies
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[19]removed: in the data assimilation system.

also introduce correlations. Liu and Rabier (2003) have shown that the assimilation can lead to sub-optimal analysis errors when [..[20]]observation-error correlations are neglected.

The weight given to the observation in the [..[21]]assimilation process is determined by its error covariance matrix $\mathbf{R}$. Therefore, its estimation plays a crucial role in the assimilation results. While most chemical assimilation systems assume the observation error to be uncorrelated, some Numerical Weather Prediction (NWP) systems have estimated non-diagonal [..[22]]observation-error covariances for satellite instruments such as Atmospheric Infrared Sounder (Garand et al., 2007; Bormann et al., 2010), IASI [..[23]](Stewart et al., 2009; Bormann et al., 2010; Weston et al., 2014; Campbell et al., 2017) and the Spinning Enhanced Visible and Infrared Imager (Waller et al., 2016). The results found in the literature for the meteorological applications incite us to account for a correlated observation error for the chemical assimilation system as well. Indeed, the studies mentioned above [..[24]]show that the inter-channel observation errors are correlated and taking such correlated errors into account in the assimilation leads to improved analysis accuracy. [..[25]]Additionally, Emili et al. (2019) has highlighted some issues when assimilating radiances in a [..[26]]chemistry transport model (increase of the ozone analysis errors compared to the control simulation at some specific altitudes), which might be due to too simplistic observation errors. The main objective of this study is, thus, to [..[27]]improve the ozone analysis accuracy within a chemistry transport model, by the mean of using more realistic observation-error covariances for IASI ozone-sensitive channels[..[28]].

The estimation of $\mathbf{R}$ is not straightforward, but a number of statistical methods are already evaluated in the literature. [..[29]]Desroziers et al. (2005) have proposed an estimation based on the observation and analysis residual statistics. By assuming gaussian errors and no correlations between observation and background errors, the error covariance matrix is provided by the statistical average of observation-minus-background times the observation-minus-analysis residuals. This method has been used in many studies to estimate the observation errors and inter-channel error correlations [..[30]](Garand et al., 2007; Weston et al., 2014; Bormann et al., 2016; Tabeart et al., 2020; Coopmann et al., 2020).

In the present work, we estimate observation errors and their inter-channel correlations for IASI using Desroziers [..[31]]method. We evaluate, then, their impact on ozone assimilation in a [..[32]]chemistry transport model (MOCAGE). The paper is organized as follows. The [..[33]]chemistry transport model, the radiative transfer model, the assimilation algorithm, the data, and the experimental framework are described in section 2. The estimation of $\mathbf{R}$ is discussed in section 3. Then, the impact on

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[28]removed: and to evaluate their impact on the analysis accuracy
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data assimilation is reported in section [..[34]]4 and validation against independent data is discussed in section 5. Finally, the summary and conclusions are given in the last section.

2 Methods and data

2.1 Methods

2.1.1 [..[35]]Chemistry Transport Model

MOCAGE (MOdèle de Chimie Atmosphérique à Grande Echelle) is the [..[36]]Chemistry Transport Model (CTM) used in this study. It is a three-dimensional CTM providing the space and time evolution of the chemical composition of the troposphere and the stratosphere. Developed by Centre National de Recherches Météorologiques (CNRM) at Météo France (Josse et al., 2004), it was used for a large number of applications such as satellite ozone assimilation (Massart et al., 2009; Emili et al., 2014), climate (Teyssèdre et al., 2007) and air quality (Martet et al., 2009). MOCAGE provides a number of optional configurations with varying domains, geometries and resolutions, as well as multiple chemical and physical parametrization packages.

A global configuration with a horizontal resolution of 2° and 60 hybrid levels from the surface to 0.1 hPa was used. The vertical resolution goes from about 100 m in the boundary layer, to about 500 m in the free troposphere and to almost 2 km in the upper stratosphere. MOCAGE is [..[37]]forced by meteorological fields from numerical weather prediction models such as Météo-France global model ARPEGE (Action de Recherche Petite Echelle Grande Echelle, (Courtier et al., 1991)), limited area model AROME (Application de la Recherche à l'Opérationnel à Méso-Echelle), and ECMWF NWP model and assimilation system (Integrated Forecast System, IFS) for air quality predictions and ARPEGE-Climat (Déqué et al., 1994) for climate simulations. In our study, the ECMWF IFS meteorological forecasts fields are used. For the chemical scheme, we adopted RACMOBUS which bundle the stratospheric scheme (Lefèvre et al., 1994) and the tropospheric scheme (Stockwell et al., 1997) including about 100 species and 300 chemical reactions.

2.1.2 Radiative Transfer Model

Remote sensing instruments measure, within a certain wavelength range, the intensity of electromagnetic radiation passing through the atmosphere (radiances). Radiative transfer models are used to simulate the radiation measured by the satellite as a function of atmospheric state, to be able to compare the model state to the observed radiances.

In our study, IASI radiances are simulated using the radiative transfer model RTTOV (Radiative Transfer for TOVS), which was developed initially for the TOVS instrument [..[38]](Saunders et al., 2018). Giving an atmospheric profile of temperature, water vapour and, optionally, trace gases, aerosols and hydrometeors, together with surface parameters and a viewing

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geometry, RTTOV simulates radiances in the infrared and microwave spectrum. For IASI, it can reproduce radiances with an accuracy of less than 0.1 K (Matricardi, 2009). In this paper, we use the same version used by Emili et al. (2019), i.e. version 11.3 (Saunders et al., 2013). The radiative transfer computations are performed in clear-sky conditions and aerosols were neglected. The surface skin temperature, 2 m temperature, 2 m pressure, and 10 m wind vector are taken from IFS forecasts. The land surface emissivity is based on the RTTOV monthly TIR emissivity atlas (Borbas and Ruston, 2010) and the Infrared Surface Emissivity Model (ISEM) (Sherlock, 1999) is used over the sea. Other chemical variables (CO_2, CH_4, CO, N_2O) were set to the reference profiles of RTTOV.

2.1.3 Assimilation algorithm

The variational data assimilation system of MOCAGE was developed jointly by CERFACS and Météo France in the framework of the European project ASSET (ASSimilation for Envisat data) (Lahoz et al., 2007). It has been used in several studies such as chemical data assimilation research (Emili et al., 2014; Massart et al., 2009), [..[39]]aerosol data assimilation (Sič et al., 2015) and tropospheric-stratospheric exchange using data assimilation (El Amraoui et al., 2010). The MOCAGE data assimilation system is flexible and allows multiple assimilation options, for example, the choice of the variational method (3D-Var, 4D-Var), the representation of the [..[40]]background-error covariance, and the type of observation assimilated. It is also used to produce operational air quality analyses for the European Project CAMS (Marécal et al., 2015).

The [..[41]]background-error covariance matrix is divided into two distinct parts, the diagonal matrix of the standard deviations and the correlation matrix. The latter, allowing to spatially smooth the assimilation increments, is modeled through a diffusion operator (Weaver and Courtier, 2001).

[..[42]]

The 3D-Var implementation has been used with hourly assimilation windows. The variational cost function is minimized using the BFGS (Broyden– Fletcher–Goldfarb–Shanno) algorithm (Liu and Nocedal, 1989). The system is preconditioned with the square root of the $\mathbf{B}$-matrix. The control vector includes only the Skin Surface Temperature (SST) and the ozone.

As we mentioned before, the aim of this work is to evaluate the impact of the estimated [..[43]]observation-error covariances on the ozone analysis. Hence, in order to be able to compare our results to those that have been already discussed and validated, we kept exactly the same configurations as those used in Emili et al. (2019) in terms of model, radiative transfer, and assimilation algorithm parameters. The summary of these configurations is [..[44]]given in table 1.

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[42]removed: In the data assimilation system of MOCAGE, the observation error covariance matrix can be read from the data file previously defined. In the case of diagonal matrix, the variances can be calculated as a percentage of the observation values.

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[..[45]][..[46]][..[47]][..[48]][..[49]][..[50]][..[51]][..[52]][..[53]][..[54]][..[55]][..[56]][..[57]][..[58]][..[59]][..[60]][..[61]][..[62]][..[63]][..[64]][..[65]][..[66]] [..[67]]

2.2 Data

2.2.1 IASI

IASI is one of the instruments operating onboard the polar-orbiting satellite Metop-A, B and C launched by the European organization for the Exploitation of Meteorological Satellites (EUMETSAT). It is based on Fourier Transform Spectrometer (FTS) and measures the spectrum emitted by the Earth-atmosphere system in the spectral range between 645 and 2760 cm^{-1} (3.62 and 15.5 µm) with a resolution of 0.5 cm^{-1} after apodization, with a spectral sampling of 0.25 cm^{-1}. IASI scans the Earth up to an angle of 48.3° on both sides of the satellite track. The cross-track is observed in 30 successive elementary fields of view, each composed of 4 instantaneous fields of view corresponding to a 12 km of diameter footprint on the ground (Clerbaux et al., 2009). The swath width on the ground is 2200 km which provides global Earth coverage twice a day. The measurements provide information on atmospheric chemistry compounds such as [..[68]]O_3, surface properties (Skin Surface Temperature SST), and meteorological profiles (humidity and temperature).

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 O_3 retrievals (Dufour et al., 2012; Emili et al., 2019). L1c data have been downloaded from the EUMETSAT Earth Observation data portal (https://eoportal.eumetsat.int, last access: 16 July 2019) in

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[52]removed: 980-1100 cm^{-1}
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[54]removed: Both Desroziers statistics and the setup of Emili et al. (2019)
[55]removed: Control vector
[56]removed: O_3 and SST
[57]removed: O_3 background error covariance
[58]removed: 3-D-hourly (standard deviation), parameterized (correlations)
[59]removed: SST prior information
[60]removed: ECMWF IFS analysis
[61]removed: SST background error standard deviation
[62]removed: 4 °C
[63]removed: T, H_2O fields
[64]removed: ECMWF IFS analysis
[65]removed: IR Emissivity
[66]removed: TIR atlas emissivity over land and ISEM model over sea
[67]removed: Table 1 : Summary of the the configuration of MOCAGE assimilation system.
[68]removed: O3

NETCDF format. Data files also contain the co-located land/sea mask and cloud fraction values, obtained from the Advanced Very High-Resolution Radiometer (AVHRR) measurements, also on board [..[69]]MetopA.

2.2.2 MLS

The Microwave Limb Sounder (MLS) provides vertical profiles of several chemical components, by measuring the microwave thermal emission from the limb of Earth atmosphere (Waters et al., 2006). More than 2500 vertical profiles are observed daily, including trace gases with a vertical resolution of approximately 3 km. Several studies benefited from MLS products, notably the ozone profiles in assimilation experiments (Emili et al., 2014; Massart et al., 2009), thanks to its low bias in the stratosphere (<5%) (Froidevaux et al., 2008).

In our study, we use the ozone profiles retrieved from MLS (V4.2 Products) as independent data to validate our results. The data have been downloaded from the Goddard Earth Sciences Data and Information Services Center (GES DISC) web portal (https://disc.gsfc.nasa.gov).

2.2.3 OMI

The Ozone Monitoring Instrument (OMI) is a nadir-viewing, ultraviolet–visible (UV-VIS) spectrometer (Levelt et al., 2018). It provides complete global maps of total column ozone on a daily basis. The OMI ozone data record starts in October 2004, shortly after the launch of Aura (McPeters et al., 2015). The total column averaged over the month of the study (July 2010), resulting from the OMI-TOMS version 8 algorithm (Bhartia, 2002), is used here to validate the results of the assimilation experiments.

2.2.4 Ozonesondes

Ozonesondes are in situ instruments carried by a radiosonde transmitting continuously the measurements as [..[70]]it ascends. The profiles of [..[71]]O_3 are provided up to an altitude that often exceeds 30 km (Jiang et al., 2007) with a vertical resolution of 150-200 m. They have been used for several applications such as validating satellite products (Jiang et al., 2007). In our study, vertical profiles of ozone, collected and distributed by the Word Ozone Ultraviolet Radiation Data Centre (http://www.woudc.org, last access), are used to validate the model simulations.

2.3 Setup of the numerical experiments

The main purpose of this study is to estimate the IASI [..[72]]observation-error covariances and verify its impact on the quality of the ozone assimilation results. [..[73]]The setup of the experiment in terms of the period of the study, the model

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configuration, the choice of assimilated observations, and of the [..[74]]background-error covariance matrix is reported in table 1 . The observation-error covariance matrix will be discussed in the section of the results (Section 3).

The model was initialized with a zonal climatology and the spin-up time used is one month (June 2010). Then, our simulations were performed for the month of July 2010. The [..[75]]ozone forecast-error standard deviation was assumed to be proportional to the ozone concentration. In fact, Emili et al. (2019) have evaluated the standard deviation of the free model simulation against independent data (profiles from ozonesondes and MLS), and found [..[76]]a small free [..[77]]forecast-error in the stratosphere, larger error in the free troposphere and highest error close to the tropopause. [..[78]]This strategy was adopted previously by many studies (Emili et al., 2014; Peiro et al., 2018; Emili et al., 2019). Emili et al. (2014) and Peiro et al. (2018) have used a percentage of 15% in the troposphere and 5% in the stratosphere. In this study, we have adopted a detailed chemical scheme (discussed in section 2.1.1). This scheme was shown to reduce the model bias compared to scheme used in Emili et al. (2014) and Peiro et al. (2018) (see Figure 4 in Emili et al. (2019)). Hence, we chose the same background error as in Emili et al. (2019) : 2% [..[79]]of the O_3 profile above 50hPa and 10% below[..[80]]. An important reason to keep the background errors similar to the setup of Emili et al. (2019) is also that we wanted to examine here exclusively the impact of $\mathbf{R}$, as already reminded in the introduction and in the conclusion.

The [..[81]]ozone background-error covariance matrix is split into a diagonal matrix filled with the standard deviation and correlation matrix modeled using a diffusion operator. The correlation, characterized by the length-scale, spreads the assimilation increments in space. [..[82]]The configurations of horizontal and vertical length scales [..[83]]are described in table 1 .

The same preprocessing described in Emili et al. (2019) has been applied to our data before their use in the assimilation system. In order to avoid any contamination from clouds, data were filtered using a cloud mask and only pixels with cloud fraction less than or equal to 1 % were kept. The cloud fraction values are obtained from the AVHRR measurements onboard [..[84]]MetopA. Since the spatial resolution of MOCAGE is coarser than the pixel size, the number of ground pixels was reduced by thinning the data using a grid of 1° x 1° of resolution and only keeping the first pixel that falls in every two grid boxes. A dynamical rejection of observations - with a threshold of 12 % - based on the relative differences between simulated and measured values with respect to simulated values was considered. Some channels affected by H_2O absorption (1008-

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[78] removed: The background standard deviation was , thus, taken equal to

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[80] removed: to mimic the validation's behavior. Similar choices were employed in (Massart et al., 2012; Peiro et al., 2018)

[81] removed: background error

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[83] removed: as in (Emili et al., 2019)

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1019,1028-1030, 1064-1067,1072-1076,1089-1092 cm^{-1}) were removed. Pixels affected by aerosols are detected and then removed using the index based on V-shaped sand signature as discussed in Emili et al. (2019).

Parameter	Configuration in the assimilation system
Period of the study	July 2010
Assimilation algorithm	Hourly 3D-Var
Radiative transfer model	RTTOV v11.3
Spectral window	980-1100 cm $^{-1}$ of IASI from MetopA
Ozone background	Hourly 3D forecasts of MOCAGE
SST prior information	ECMWF IFS forecasts
Control vector	O_3 and SST
T, H_2O fields	ECMWF IFS forecasts
IR Emissivity	TIR atlas emissivity over land and ISEM model over sea
Observation-error covariance	Both Desroziers method and the setup of Emili et al. (2019)
SST background-error standard deviation	4 °C
O_3 Background error	Vertically variable and computed as % of the background profile (using a value of 2% above 50 hPa and 10 % below)
O_3 Background-error zonal correlation	Exponential with a length scale set to 200 Km and reduced towards the pole to account for the increasing zonal resolution of the regular latitude-longitude grid.
O_3 Background meridional error correlation	Exponential with a length scale set to 200 Km.
O_3 Background-error vertical correlation	Exponential with a length scale set to 1 grid point (vertical level).

Table 1 : Summary of the the configuration of MOCAGE assimilation system.

3 R estimation

3.1 Desroziers diagnostics

The observations used in the assimilation system could have a margin of error. We can identify two types of errors, systematic and random errors. The systematic error is ordinarily corrected before the data assimilation process. [..[85]]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

[85]removed: Nevertheless, this correction should not account for the model bias, which

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 [..[86]]analyses to unrealistic values that might be introduced by the model bias. In our case, we [..[87]]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 and McNally (2010), have used the channel 1585 as an anchor in the [..[88]]assimilation of ozone for NWP. Dragani and Mcnally (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 (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 [..[89]]was adopted in many chemical analyses' studies before (e.g. Massart et al. (2012); Peiro et al. (2018); Emili et al. (2019)).

Random errors can arise from the measurements (e.g. instrumental error), forward model, representativeness error (e.g. difference between point measurements and model representation), or quality control error (e.g. error due to the cloud detection scheme missing some clouds within clear sky only assimilation). These types of errors should be accounted by the [..[90]]observation-error covariances matrix $\mathbf{R}$. According to [..[91]]Weston et al. (2014), the instrument noise could be assumed to be uncorrelated. However, the IASI measurements are apodized, which may introduce correlations between neighboring channels, particularly in our case where we are assimilating a subset of adjacent channels. The radiative transfer model may also introduce correlations between channels. The statistics of error from the instruments noise are known, while the characteristics of other sources of error are not yet well understood.

In this paper, we estimate the total error using the statistical approach introduced by [..[92]]Desroziers et al. (2005) .

[86] removed: analysis

[87] removed: are assimilating troposphere and stratosphere data,

[88] removed: missing of an anchor instrument in the top of stratosphere led us to

[89] removed: , in fact, is adopted in most chemical analyses(Emili et al., 2019; Massart et al., 2012).

[90] removed: observation error

[91] removed: (**?**)

[92] removed: **?**

$$[..^{93}\ ]\mathbf{R} = \mathbf{E}[(\mathbf{y} - \mathbf{H}(\mathbf{x}_a))(\mathbf{y} - \mathbf{H}(\mathbf{x}_b))^T]$$

Where $\mathbf{x}_a$ is the analysis state vector, $\mathbf{x}_b$ is the background state vector, $\mathbf{y}$ is the vector of observations and $\mathbf{H}$ is the observation operator that computes model counterpart in the observation space.

This method has been used to estimate observation errors and inter-channel error correlations [..[94]](Stewart et al., 2009; Bormann et al., 2016; Tabeart et al., 2020; Coopmann et al., 2020). It can potentially provide information on imperfectly known observation and background-error statistics with a nearly cost-free computation [..[95]](Desroziers et al., 2005). However, this approach assumes that the $\mathbf{R}$ and $\mathbf{B}$ matrices used to produce the analysis are exactly correct, which may not be always the case. Furthermore, Desroziers diagnostics compute the total covariances, more efforts are needed to understand and distinguish the sources of the error.

3.2 Error results

The Desroziers [..[96]]method was computed on the output of a 3D-Var experiment using a diagonal matrix $\mathbf{R}$ (with a standard deviation of 0.7 $\mathrm{mWm^{-2}sr^{-1}cm}$ as in [..[97]]Emili et al. (2019)). The diagnosed $\mathbf{R}$ could not be used directly in the assimilation system. In fact, the estimated matrix was asymmetric and not positive definite. [..[98]]Similar unrealistic features in the diagnosed covariance matrices were encountered in [..[99]](Stewart et al., 2014; Weston et al., 2014) where an artificial inflation of observation errors was [..[100]]applied. $\mathbf{R}$ needs to be a valid covariance matrix before being used in the 3D-Var assimilation system. Therefore, we first symmetrize the estimated matrix by taking the mean of the original matrix and its transpose. Then we impose the negative eigenvalues to be equal to the smallest positive eigenvalue [..[101]]as in (Weston et al., 2014; Tabeart et al., 2020). Another method which consists of increasing all eigenvalues of $\mathbf{R}$ by the same amount was tested here to recondition the estimated matrix. We favoured the first method since the standard deviation and the correlation values remain closer to the initially estimated quantities.

[..[102]]Using outputs (analyses and forecasts) derived from 3D-Var experiment that uses a diagonal $\mathbf{R}$[..[103]]-matrix (called hereafter 1^{st} 3D-Var experiment) in the estimation process might have an impact on the diagnosed $\mathbf{R}$-matrix. The matrix derived using these outputs is called hereafter 1^{st} estimation. We performed another 3D-Var experiment [..[104]](2^{nd} 3D-Var experiment) using the 1^{st} estimation. The outputs (analyses and forecasts) of this experiment (2^{nd} 3D-

[93] removed: $\mathbf{R} = \mathbf{E}[(\mathbf{y} - \mathbf{H}(\mathbf{x}_a))(\mathbf{y} - \mathbf{H}(\mathbf{x}_b))]$

[94] removed: (Stewart et al., 2009)

[95] removed: (?)

[96] removed: statistics were

[97] removed: (Emili et al., 2019)

[98] removed: This might result from the violation of one of the assumptions of Desroziers statistics stipulating that the $\mathbf{B}$ and $\mathbf{R}$ matrices used to produce the analysis were correctly defined.

[99] removed: (Stewart et al., 2014; ?)

[100] removed: assumed

[101] removed: (?). An other method was tested here to recondition the estimated matrix, the one called *ridge regression* (??)

[102] removed: In order to evaluate the impact of starting from an experiment based on

[103] removed: matrix, we performed a second

[104] removed: using the diagnosed and modified matrix. Then, we used these new forecasts and analyses to estimate again

Var experiment) were used to estimate another $\mathbf{R}$[..[105]]-matrix called 2^{nd} estimation. The standard deviation of the 2^{nd} estimation is larger than that of the 1^{st} estimation (not shown). The same goes for [..[106]]correlations (not shown). It should be noted that the [..[107]]2^{nd} estimation was positive definite, unlike the [..[108]]1^{st} estimation where some unrealistic features were encountered. We have followed the same process to [..[109]]further estimate two other matrices ([..[110]]3^{rd} and 4^{th} estimation). The differences [..[111]]of the estimations in terms of standard deviation and correlations became smaller as we reestimate the matrices[..[112]], suggesting a sort of convergence of the estimation. [..[113]]We have adopted the 2^{nd} estimation for the results shown in this work. The reason for this choice will be [..[114]]discussed later (section 5.2).

Figure 1 presents the standard deviation diagnosed [..[115]]using the Desroziers approach (solid black line) and that used in Emili et al. (2019) (dotted blue line). The latter was set equal to 0.7 mW m^{-2} sr^{-1}cm for all channels, which is a common setting for most IASI [..[116]]O_3 retrievals (Dufour et al., 2012). At first glance, we note that the standard deviation used in previous studies is highly underestimated for the SST sensitive channels and overestimated for some [..[117]]ozone-sensitive channels (around 1040 and 1050 cm $^{-1}$). The diagnosed standard deviation increases to reach 2 mW m^{-2} sr^{-1} cm for SST sensitive channels (the first and the last twenty channels of the band (980-1000 cm^{-1} and 1080-1100 cm^{-1}) and the channels between 1040 and 1045 cm^{-1}) and varies from 0.2 to 1.4 mW m^{-2} sr^{-1} cm for the [..[118]]ozone-sensitive channels. The radiance values for the observations are greater for the SST channels than those of the ozone. The same goes for the corresponding background and the analysis values. Since these diagnostics are based on observation, background and analysis residuals, a larger standard deviation for the SST channels than for ozone channels might be expected. We remarked that the estimated standard deviation is proportional to the radiance value (either the observation, the background, or the analysis value), which gives a relative standard deviation of about 2.5 % of the average radiances values for the entire spectral window (not shown).

The IASI instrumental error is provided by the CNES (Centre National d'Etudes Spatiales), taking into account different known effects such as flight homogeneity and apodization effect (Le Barbier Laura, personal communication, September 18, 2019). The instrumental error covariance matrix is computed as described in (Serio et al., 2020). This error remains smaller

[105] removed: with the Desroziers diagnostics. The resulted standard deviation was significantly greater than the one estimated from the experiment that use a diagonal matrix

[106] removed: the

[107] removed: re-estimated matrix

[108] removed: first

[109] removed: reestimate

[110] removed: introduce, each time, the estimated matrix into the assimilation system in order to have analyses that we use to re-estimate a new matrix

[111] removed: between

[112] removed: (not shown)

[113] removed: All matrices and simulations discussed in this paper are based on the second estimation (which uses the analyses derived from the experiment using the first conditioned estimation)

[114] removed: rediscussed

[115] removed: (second estimation)

[116] removed: O3

[117] removed: ozone sensitive

[118] removed: ozone sensitive

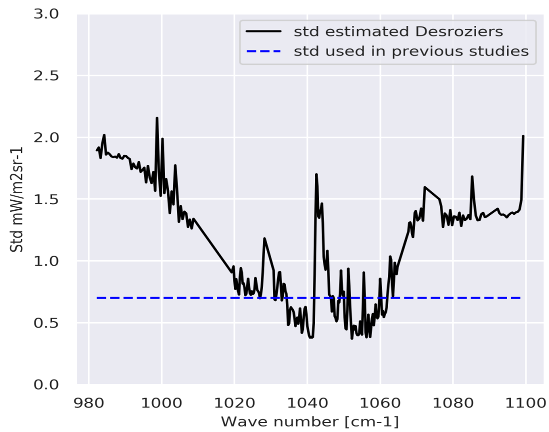

Figure 1. Standard deviation estimated using (back solid line) and that used in the previous studies (dotted blue line) (Emili et al., 2019) .

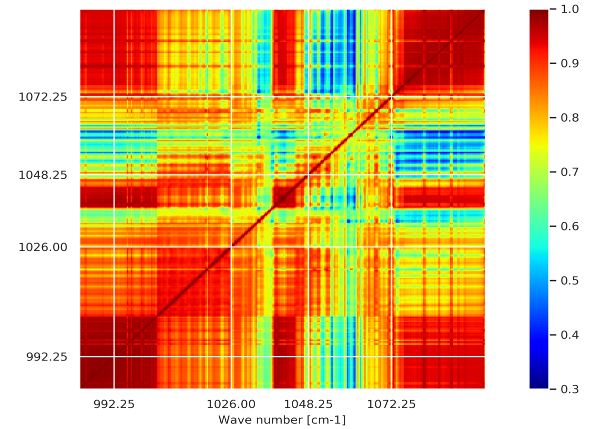

Figure 2. Correlation matrix estimated using the Desroziers method.

([..[119]]about 0.2 mW m^{-2} sr^{-1}cm) than that used in the previous studies (0.7 mW m^{-2} sr^{-1}cm). Then, the [..[120]]large estimated standard deviation noticed in our estimation might be due to the radiative transfer inputs error.

To investigate the off-diagonal part of $\mathbf{R}$ we present the diagnosed correlation matrix in [..[121]]Figure 2. The results show high correlations between the majority of the channels (larger than 0.4). In particular, a very high correlation is observed among SST sensitive channels (around 0.9 to 1). The regions of, relatively, lower correlation (around 0.4 to 0.7) represent the ozone channels correlations and cross correlation between ozone and SST sensitive channels.

The high correlation found here was expected since previous studies have highlighted the same behaviour in this spectral region [..[122]](Bormann et al., 2010; Stewart et al., 2014; Bormann et al., 2016). In fact, the use of the same radiative transfer model for all channels may introduce similar errors among these channels.

The diagnostic discussed above is based on a global estimation, without any distinction between the type of the surface (land or sea) nor the time of the observation (day or night). Since the emissivity varies according to the type of the surface, and the skin temperature is strongly driven by the sun radiation, we evaluated $\mathbf{R}$ taking these differences into account.

[119] removed: maximum of 1.06e^{-6}

[120] removed: important

[121] removed: figure

[122] removed: (Bormann et al., 2010; Stewart et al., 2014; **?**)

[..[123]]In terms of standard deviation, the error over land reveals large values for the SST [..[124]]sensitive-channels in comparison with that estimated over the sea which, in turn, reproduces a slightly different error in comparison with the global estimation (not shown). The two surfaces introduce also a slightly different error regarding the ozone band. [..[125]]

[..[126]]

The same behaviour as the global estimation is reproduced when the statistics were performed from the data measured separately from the day and from the night. [..[127]]The variability in terms of correlations is more pronounced when the surface type is considered than in the [..[128]]

[..[129]]

[..[130]]case of the observation time. The difference between the correlations estimated using all observations and pixels over the sea surface varies between 0 % and 40 % for the majority of the channels with values that can reach 60%. These differences are located around 1035 and 1060 cm^{-1} which correspond to the regions of low correlations [..[131]]

[..[132]](not shown).

[..[133]]The separate treatment of land/sea covariance matrices did not yield significant differences in terms of [..[134]]assimilation results comparing with the use of global estimation. Hence, we have adopted the global estimation [..[135]]in our study. The rationale for this choice will be given during the discussion of the validation results (section 5.2).

[123]removed: Figure 3 shows the difference between the standard deviationestimated using data localized over the sea and land separately. The

[124]removed: sensitive channels

[125]removed: Since the number of observations over the sea is greater than over the land (almost 65 %), the global estimation is dragged down to be close to the sea estimation. To explain the large difference in the region of SST sensitivity channels over the land, the surface properties need to be discussed.

[126]removed: The surface emissivity varies with vegetation, soil moisture, composition, and roughness (**?**) with values between 0.65 and 1 in the thermal infrared range. Low values are found in deserts and high values over dense vegetation and water surface (**?**). The variability of the soil type over land in comparison with the sea that is homogenous can lead to a larger standard deviation over land than over sea. The second parameter that impacts the electromagnetic spectrum measured by the infrared sensor is the surface temperature. Its variability is larger over land than over sea, which also contributes to the difference of the standard deviation between land and sea. On the other hand, these differences are larger for the SST channels than for the ozone channels. This might be explained by the direct link between the radiance signal and the surface signal in the SST bands especially since the observation and the estimated standard deviation were found proportional (mentioned above).

[127]removed: Figure 4 shows small differences in the SST channels sensitivity band (between 1080 and 1100 cm^{-1}), whereas the curves take approximately the same values for the rest of

[128]removed: channels.

[129]removed:

[130]removed: The data assimilation process uses the full error covariance matrix in order to minimize the cost function. Hence, we discuss now the differences between the correlation matrices taking the time (day/night) and the surface type (sea/land) of the observations into account. Figure 5 shows the correlation matrix estimated using all observations (a), only pixels over sea (b), only pixels over land (c), the difference (in %) between the global and sea matrix (divided by the global matrix) (d), and the difference (in %) between global and land matrix (divided by the global matrix) (e). According to the figures (a), (b), and (c), the differences between the three matrices differences may appear small. However, by looking at the figures (d) and (e), the

[131]removed: .

[132]removed: The same comparison was held separating observations from the day and from the night: significantly smaller differences compared to the land and sea separation were noticed

[133]removed: In conclusion, the variability

[134]removed: correlations is more pronounced when the surface type is considered than in the case of the observation time. For the latter, the smaller differences found for the standard deviation (Fig. 4) are also found for correlations. For the follow-up assimilation experiments, we

[135]removed: and neglected the effect of the time and surface of the observations

[..[136]][..[137]]

4 Assimilation results

4.1 Ozone fields

In this section, we discuss the impact of the [..[138]]observation-error covariances estimated previously on the ozone analysis. To this end, three experiments for the month of July 2010 were carried out:

i). [..[139]]model run without data assimilation called hereafter the free run (or Control), and noted in the rest of this paper ControlExp.

ii). [..[140]]3D-Var assimilation of IASI radiances using a diagonal [..[141]]observation-error covariance matrix (as in Emili et al. (2019)). It will be referred here by [..[142]]RdiagExp.

iii). [..[143]]3D-Var assimilation of IASI radiances using a full matrix estimated with the Desroziers diagnostic noted hereafter by RfullExp.

The first experiment (ControlExp) was run to evaluate the benefit of the assimilation experiments and to quantify the improvements of each of the two analyses when they are validated against independent data. The same setup of Emili et al. (2019) was adopted for [..[144]]RdiagExp, which was taken as a reference to characterize the impact of accounting for the estimated $\mathbf{R}$ in the third simulation (RfullExp).

Figure [..[145]]3 shows the difference between the zonal average of the ozone analysis from the two assimilation experiments in units of parts per billion volume (ppbv). The zonal values were averaged over the month of the study before performing the difference. The impact of the estimated $\mathbf{R}$ varies with latitude. It varies also with the height, adding or reducing the amount of ozone. Overall, the estimated $\mathbf{R}$ reduces the amount of ozone in the high latitudes of the free troposphere and the tropical high stratosphere, whereas the amount is increased in the vicinity of the lower stratosphere. The maximum reduction of ozone is larger than the amount added. The amount of ozone reduction reaches 600 ppbv, whereas the increase does not exceed 300 ppbv. In high northern latitudes (30°N-90°N), a significant addition is found (300 ppbv) covering almost the whole stratosphere, in opposition to the other latitudes where the difference changes sign with altitude. On the other hand, an important reduction

[136]removed: H

[137]removed: **(a)** Correlation matrix estimated using all observations, **(b)** using pixels observed over the sea, **(c)** using pixels observed over the land, **(d)** difference (in %) between global and sea matrix (divided by the global matrix), **(e)** and the difference (in %) between global and land matrix (divided by the global matrix)

[138]removed: observation error covariance

[139]removed: a

[140]removed: A

[141]removed: observation error

[142]removed: RefExp

[143]removed: A

[144]removed: RefExp

[145]removed: 6

of ozone is observed in the tropics at 20 hPa (more than 600 ppbv). To better understand the impact of the estimated $\mathbf{R}$ we validate the results with independent data in the section of validation (section 5).

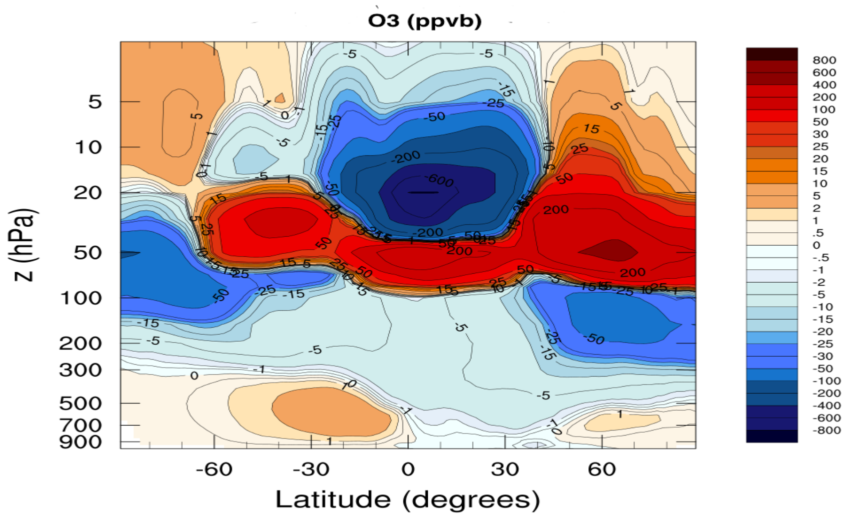

Figure 3. The difference between the zonal average of the analysis (in ppbv) from the two assimilation experiments, averaged over the month of the study (nonlinear colormap).

[..[146]]

4.2 Surface skin temperature

The assimilated spectra include both ozone and surface skin temperature sensitive channels. The IFS skin temperature was taken as a background in the assimilation process. We have computed the difference between the SST analysis and the background at the end of each assimilation experiment ([..[147]]RdiagExp and RfullExp). [..[148]]The skin temperature is physically linked to the ozone measured. In fact, the skin temperature interacts with the ambient atmosphere. An increase of SST can for example create a convective movement impacting the transport of the ozone. However, the skin temperature is given only at the observation location in this study and it is specified with values interpolated from NWP forecasts (IFS), whereas ozone is a 3D field issued from the chemistry transport model. Hence, the estimation and potential account of error

[146]removed: The emphasis, up to now, has been on the impact of the estimated observation error on the ozone analysis. We discuss in the following subsection the impact on the skin surface temperature analysis.

[147]removed: RefExp

[148]removed: Figure 7.

correlations between the two variables seems challenging in our system. In this work, we did not consider the background-error correlation that might exist between O_3 and SST.

Figure 4.a) shows the difference between the analysis of the SST given by [..[149]]RdiagExp and the IFS SST forecast whereas, [..[150]]Figure 4.b) shows the difference between the analysis of the SST given by RfullExp and the IFS SST forecast. In terms of geographical distribution, we notice that the differences are smaller through the tropics and mid-latitudes, especially over sea, when the estimated $\mathbf{R}$ was adopted. Looking at the average values, [..[151]]RdiagExp decreases the surface skin temperature by about [..[152]]0.55°C with respect to the background. The introduction of the estimated $\mathbf{R}$ decreases the difference between the SST analysis and that of IFS to almost [..[153]]-0.18°C instead of [..[154]]-0.55 °C. The standard deviation was also reduced from [..[155]]1.39 °C [..[156]]to 1.05 °C. Thus, the use of the estimated $\mathbf{R}$ lets the SST analysis stay closer to the IFS forecasts[..[157]]. However, there is an increase in difference on land using RdiagExp, mainly in Africa and South America. This increase in difference over the land seems related to the dependence of observation errors on the surface. In fact, the number of observations over the sea represents almost 70% of the total observations we have used in this study. Consequently, our SST analysis stays closer to background values (IFS forecasts) over the sea than over the land.

[149] removed: RefExp
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[151] removed: RefExp
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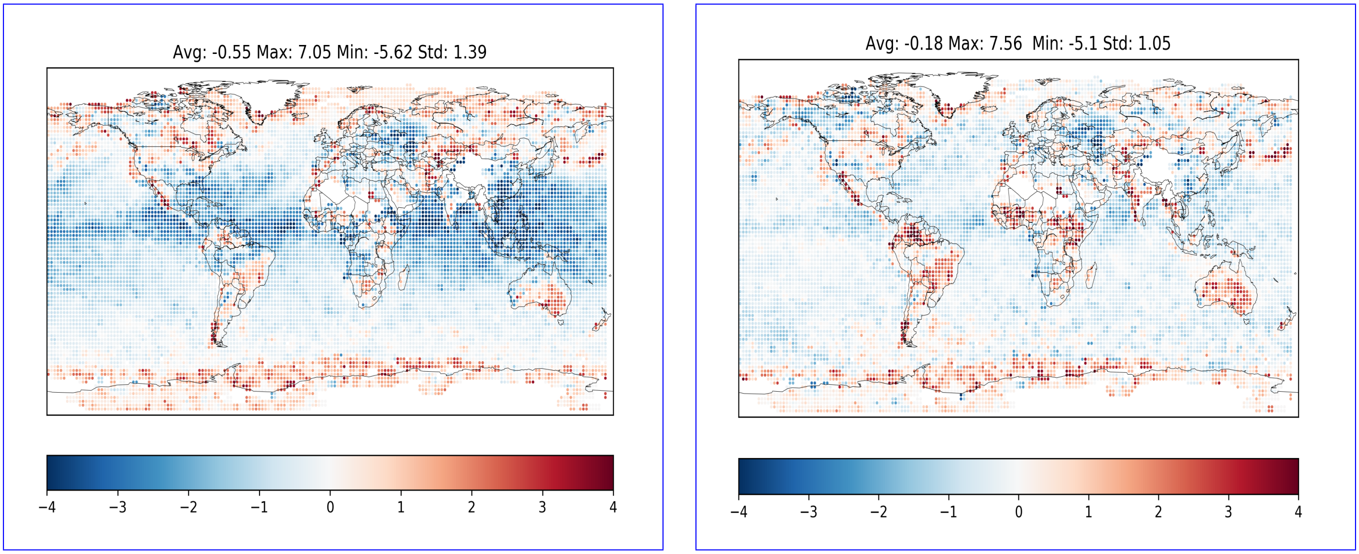
[157] removed: which are reliable data.



(a) **(b)**

Figure 4. Difference (in °C) between the IFS SST forecast and the analysis of the SST given by [..[158]]RdiagExp (with a diagonal matrix) (**a**), and that given by RfullExp (with a correlated matrix) **(b)** averaged by box of 2°.

4.3 Computational cost

In our assimilation setup, the cost function is minimized hourly. For each window, the minimizer needs to converge after a certain number of iterations. The cost of each iteration is dominated by the cost of the radiative transfer operators (tangent linear, the adjoint model) and of the [..[159]]background-error covariance operators. When the observation error was assumed to be uncorrelated ([..[160]]RdiagExp), the number of iterations needed for each hourly cycle is significantly higher than when the estimated [..[161]]observation-error covariance matrix is used. In fact, the introduction of the estimated $\mathbf{R}$ reduces the number of iterations from 150 (a fixed value to stop iterations if the convergence criteria were not achieved to save computational time) to 90 iterations in average. This means that the CPU time is reduced by more than 150% for each assimilation cycle. This improvement is highly valuable when the study is extended to longer periods. This reduction is due to the fact that the diagonal matrix is pulling much closer to the observations than the correlated matrix, which makes it harder to find a solution resulting in slower convergence. This increase of the convergence speed was encountered in the Met Office 1D-Var system (Tabeart et al., 2020) where a correlated observation matrix was introduced in the system. [..[162]]Moreover, in Tabeart et al. (2018) the matrix $\mathbf{R}$ and the [..[163]]observation-error variance appear in the expression of the condition number of the Hessian of the variational assimilation problem, indicating that these terms are important for convergence of the minimization function. [..[164]]

[159]removed: background error
[160]removed: RefExp
[161]removed: observation error
[162]removed: Furthermore
[163]removed: observation error
[164]removed: To understand which one between correlations and variance has a higher impact

In an attempt to distinguish the impact of the variance on the convergence speed [..[165]]from that of the correlations, we have performed three assimilation experiments using different $\mathbf{R}$-matrices. The first experiment (1^{st} experiment) employed $\mathbf{R}$ that was estimated from the analysis computed using a diagonal $\mathbf{R}$-matrix. The minimizer takes generally more than 100 iterations to converge. We used the analysis given by the 1^{st} experiment to estimate another $\mathbf{R}$-matrix. We have used this estimation to run another assimilation cycle (2^{nd} experiment). We have noticed that the minimizer needs about 60 iterations to converge. We have modified the $\mathbf{R}$-matrix of the 1^{st}experiment by keeping its correlations and [..[166]]replacing its standard deviation with that of [..[167]]$\mathbf{R}$ used in the 2^{nd} experiment. The resulting matrix was used to run a 3^{rd} assimilation experiment. The minimizer [..[168]]needs less than 100 iterations to converge (about 70 iterations). Actually, using the 1^{st} estimation of $\mathbf{R}$, the minimization needs more than 100 iterations to converge, whereas about 60 iterations are needed with the 2^{nd} estimation of $\mathbf{R}$. The results of the 3^{rd} experiment seem to suggest that updating the variance has a [..[169]]larger impact on the convergence speed.

5 Validation of [..[170]]O_3 analyses

5.1 Total column

Figure [..[171]]5 shows the difference of the ozone total column (in Dobson Unit (DU)) provided by OMI and that of the [..[172]]RdiagExp (a) and that of RfullExp (b). At first sight, we note smaller differences over the tropics between the OMI total column and the total column given by RfullExp in comparison with that given by the [..[173]]RdiagExp. This behaviour was expected since [..[174]]a large reduction of the amount of ozone was observed in these regions (see [..[175]]Figure 3). In the high northern latitudes, the differences were slightly increased when the estimated matrix was adopted. This is a consequence of the increase in the amount of ozone encountered in these regions in the stratosphere, compared to the amount reduced in the same region in the troposphere ([..[176]]Figure 3). On the other hand, the global mean and the standard deviation of these differences are lower in the case of using the new estimated matrix [..[177]]

(10.1 DU as a mean and 6.3 as a standard deviation when the new estimated matrix was used instead of 10.6 DU as a mean and 7.3 as a standard deviation when a diagonal matrix was used). Hence, we conclude that the estimated matrix $\mathbf{R}$ has slightly improved the results in terms of ozone total columns.

[165] removed: , we proceeded as follows: we considered an estimation that did not lead to a convergence (the first estimation) of the minimizer , we kept
[166] removed: replace
[167] removed: an estimation that leads to a convergence (the second estimation)
[168] removed: , consequently, converges allowing us to conclude that
[169] removed: higher
[170] removed: O3
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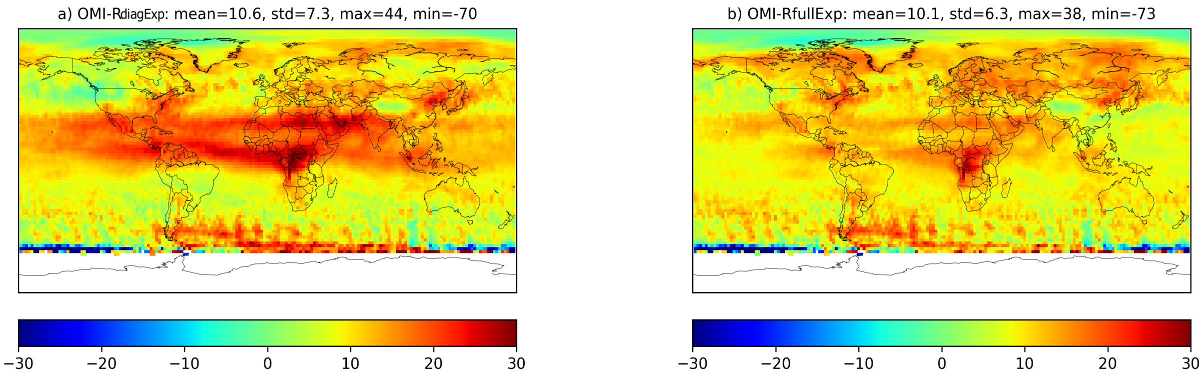

Figure 5. (a) Difference of the ozone total column (DU) provided by OMI and that of the assimilation experiment [..[178]]RdiagExp,**(b)** and that of RfullExp, averaged over the month of the study.

5.2 Vertical validation

In this section, we validate the two simulations against radiosoundings and MLS data. We use the root mean square error (RMSE) as the main statistical indicator to quantify the accuracy of the experiments.

We compute the relative (to the control simulation) difference of RMSE with respect to radiosoundings and MLS averages globally and for five different latitude bands. The difference is computed by subtracting the RMSE of each experiment from that of the control simulation. Negative values indicate an improvement of the [..[179]]O_3 profiles. It should be noted that the representativeness of the statistics given by the MLS in the stratosphere is better than that of the radiosoundings because the number of profiles provided by MLS is much higher compared to the radiosoundings ones. Consequently, higher confidence is given to the validation using the MLS data in the stratosphere.

[179]removed: O3

Figure [..[180]]6 reports the RMSE differences with respect to the radiosoundings. Considering the global RMSE (ALL), we notice that the experiment with the estimated matrix improves the results above 150 hPa, around 400 hPa and near the surface. However, it also reduces the improvement from 30% (the case of using a diagonal matrix) to 15 % in the vicinity of UTLS (100-300 hPa).

The [..[181]]issue of increasing the [..[182]]ozone analysis errors compared to the control simulation encountered in Emili et al. (2019) is especially severe in the tropics (30S-30N). The use of the estimated $\mathbf{R}$ has substantially enhanced the results in this latitude bands bringing the error from +15% to -2%. Apart from the vicinity of 50 hPa and 400 hPa, the results, in the tropics, were improved over the entire vertical profile. Regarding other latitude bands, almost the same feature of the global validation is found in the north hemisphere. The two experiments show almost the same behaviour in the southern latitudes, with a slight improvement for RfullExp in the southern high latitudes (60°S-90°S).

The MLS validation in [..[183]]Figure 7 shows almost the same behaviour reported by radiosoundings validation in the tropical stratosphere, where the reduction of error is remarkable. In the other latitude bands, MLS reports a similar behaviour of the two experiments, with some small differences in northern hemisphere.

All things considered, the introduction of the estimated $\mathbf{R}$ has globally improved the [..[184]]O_3 profiles in the stratosphere and in the free troposphere, especially in the tropics. In spite of its degradation in the vicinity of UTLS, the improvement remains always advantageous with respect to the control run.

The matrix used for this study (see Sec. 3.2) will be now discussed in this section since the decision was also based on the outcome of the assimilation experiments presented in this section. We performed sequentially three assimilation experiments using the first, second and the third estimation of $\mathbf{R}$ (Sec. 3.2). The results of validation against radiosoundings and MLS showed small differences (not shown). Therefore, to avoid the initial impact of using a diagonal matrix we have adopted the second estimation (which uses the analyses derived from the experiment using the first [..[185]]estimation). In an operational framework, we may estimate the matrix daily (weekly or monthly if the period of the study is considerably long) using the analyses of the previous day (using the analysis of the previous week or month respectively). In other words, we may use a diagonal matrix to produce analyses for the first day or spin-up period, these analyses will be used to estimate the matrix that will be used for the second day, and so on throughout the period of the study.

We have also discussed the type (sea/land) and the time (day/night) of the observations while estimating the matrices. To check the impact of these differences on the assimilation results, we ran an additional assimilation experiment using the matrix estimated considering the type of the surface of each observation (since the differences were more important than if the time of the observation was considered). Only slight differences among the results have been noticed (not shown). 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. The differences, in terms of standard deviation, of the global

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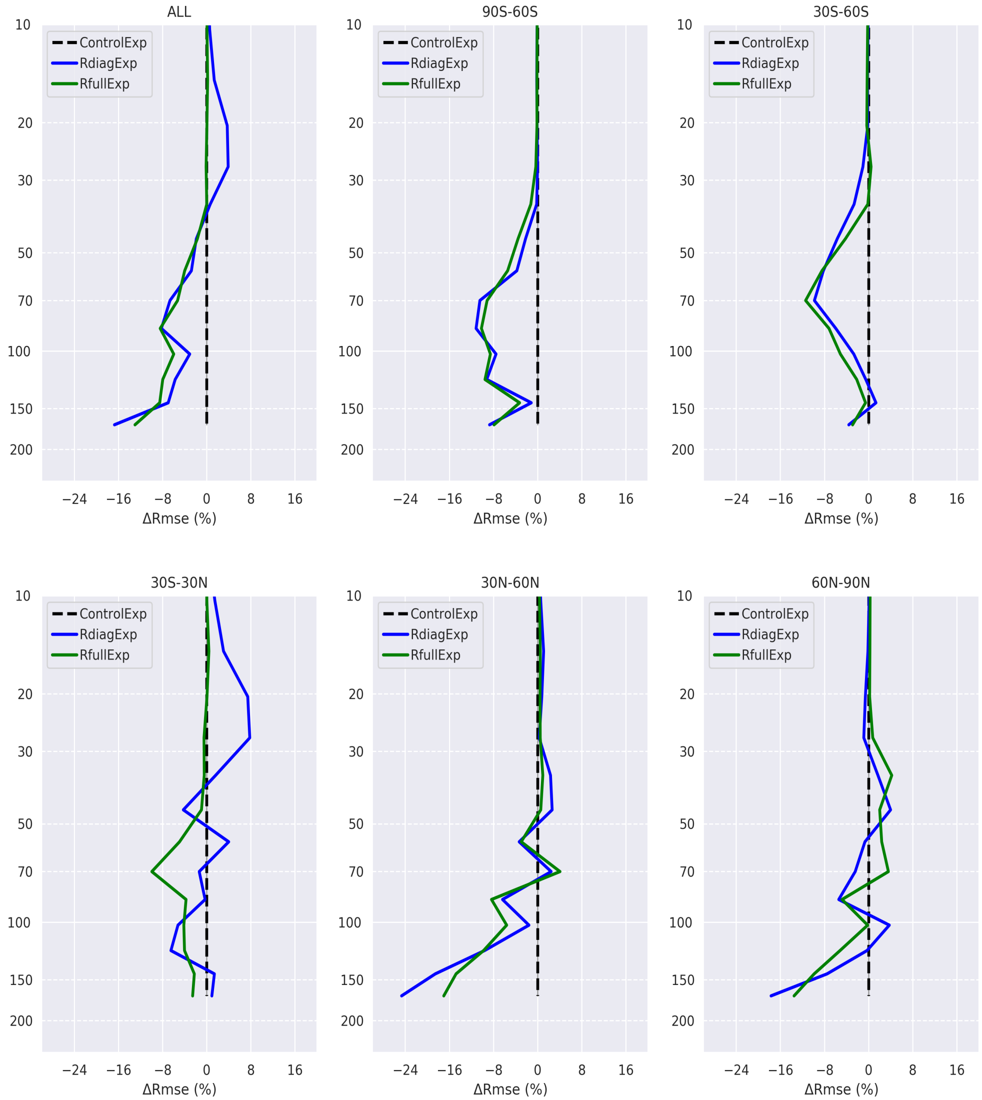

Figure 7. [..[194]]Normalized difference of the RMSE with respect to the MLS [..[195]]for the [..[196]]R estimated (green) and [..[197]]R diagonal (blue). The [..[198]]difference of the RMSE was computed by subtracting the RMSE of the [..[199]]analysis from the RMSE of the [..[200]]control of each experiment, 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. (Vertical levels are in hPa).

6 Conclusions

The correct specification of the observation error becomes a critical issue to assimilate efficiently the increasing amount of satellite data available in the recent years. We have estimated the observation errors and their inter-channel correlations for clear sky radiances from IASI [..[201]]ozone-sensitive channels. We have evaluated, then, the impact of the estimated $\mathbf{R}$ on the SST and ozone analysis within our 3D-Var assimilation system. The outcome has been compared with an assimilation experiment where the [..[202]]observation-error covariance matrix was assumed to be diagonal and the standard deviation assigned empirically like in previous studies. The results of the experiments were, then, validated against independent data: OMI, MLS and ozonesondes.

The Desroziers diagnostics were adopted here to estimate $\mathbf{R}$. The diagnostics used the analyses derived from a variational data assimilation experiment. The results have shown high correlations between the majority of the IASI spectral channels, particularly among the SST sensitive channels.

[..[203]]

Significant differences between the results of the experiments were encountered. The introduction of the estimated $\mathbf{R}$ reduces the amount of ozone in the free troposphere and in the high tropical stratosphere, whereas ozone is added in the vicinity of the lower stratosphere. [..[204]]A validation against OMI has shown that the results were closer to the observations when the estimated matrix was adopted.

The validation against MLS and ozonesondes showed that the introduction of the estimated $\mathbf{R}$ has globally improved the results in the stratosphere and in the free stratosphere especially in the tropics. In spite of a slight reduction in accuracy in the vicinity of UTLS, the improvement remains always advantageous with respect to the reference assimilation. Concerning the computational cost, the introduction of the estimated $\mathbf{R}$ significantly reduces the number of iterations needed for the minimizer to converge.

In summary, accounting for an estimated $\mathbf{R}$ improves significantly the ozone assimilation results. [..[205]]This approach might be adopted in the assimilation of other chemical species and also in [..[206]]Level 2 O_3 retrievals.

In this study, the estimation was computed without taking into account any distinction of the error sources and assuming that the observation error was unbiased. More efforts will be needed to tackle these points. It should also note that we kept the same experiment setup of Emili et al. (2019) in order to be able to quantify exclusively the impact of the $\mathbf{R}$. The [..[207]]background-error matrix was still defined using a relatively simple and empirical method. Further research might be needed to perform a

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[203] removed: The impact of the surface type (land/sea) and the time of the observation (day/night) on the estimation was evaluated. Significant differences were noticed in terms of standard deviation when the surface type was considered. However, slight differences in the results have been noticed when the matrix estimated considering the time and the type of the surface of the observation was used in the assimilation experiment. A global estimation of the matrix could finally be adopted.

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[206] removed: a chemical retrieval process framework

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better estimation of the background error. A new channel selection might also be performed to reduce the computational cost and the information redundancy of the IASI spectrum. On the other hand, all the experiments are performed in the context where aerosols are neglected and over one month. Including modelled aerosols within the radiative transfer may bring some improvements to the analyses. These aspects will be covered in future research.

Competing interests. The authors declare that they have no conflict of interest.

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[208]removed: chemical