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 (CO_2, CH_4, CO, N_2O, SO_2)? Do you use reference profiles? Which coefficient file do you take into account?*

Answer

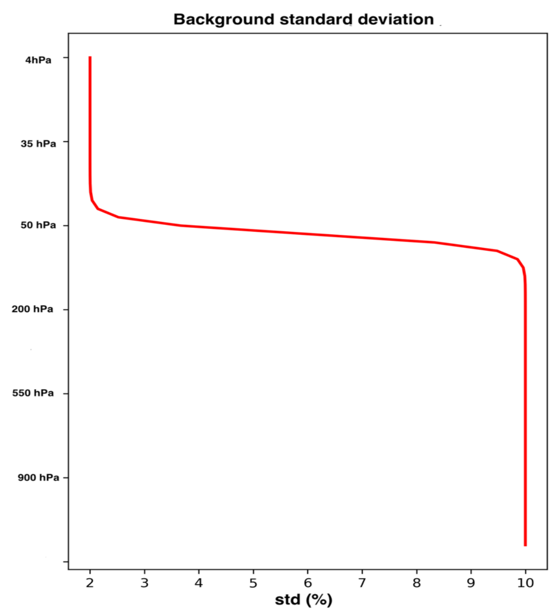
These chemical variables (CO_2, CH_4, CO, N_2O) were set to the reference profiles of RTTOV. For the coefficient file, we used the coefficients for v9 predictors computed on 101 levels.
The SO_2 was not available in RTTOV v11 (used in this study).
We have added this comment to the paper (L13 P4 of the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



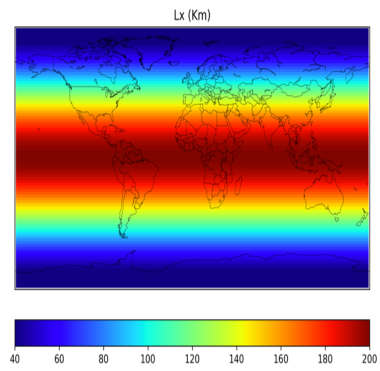


Figure 1_R1 : background-error standard deviation (square root of the diagonal of B) in % of the background profile.

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.

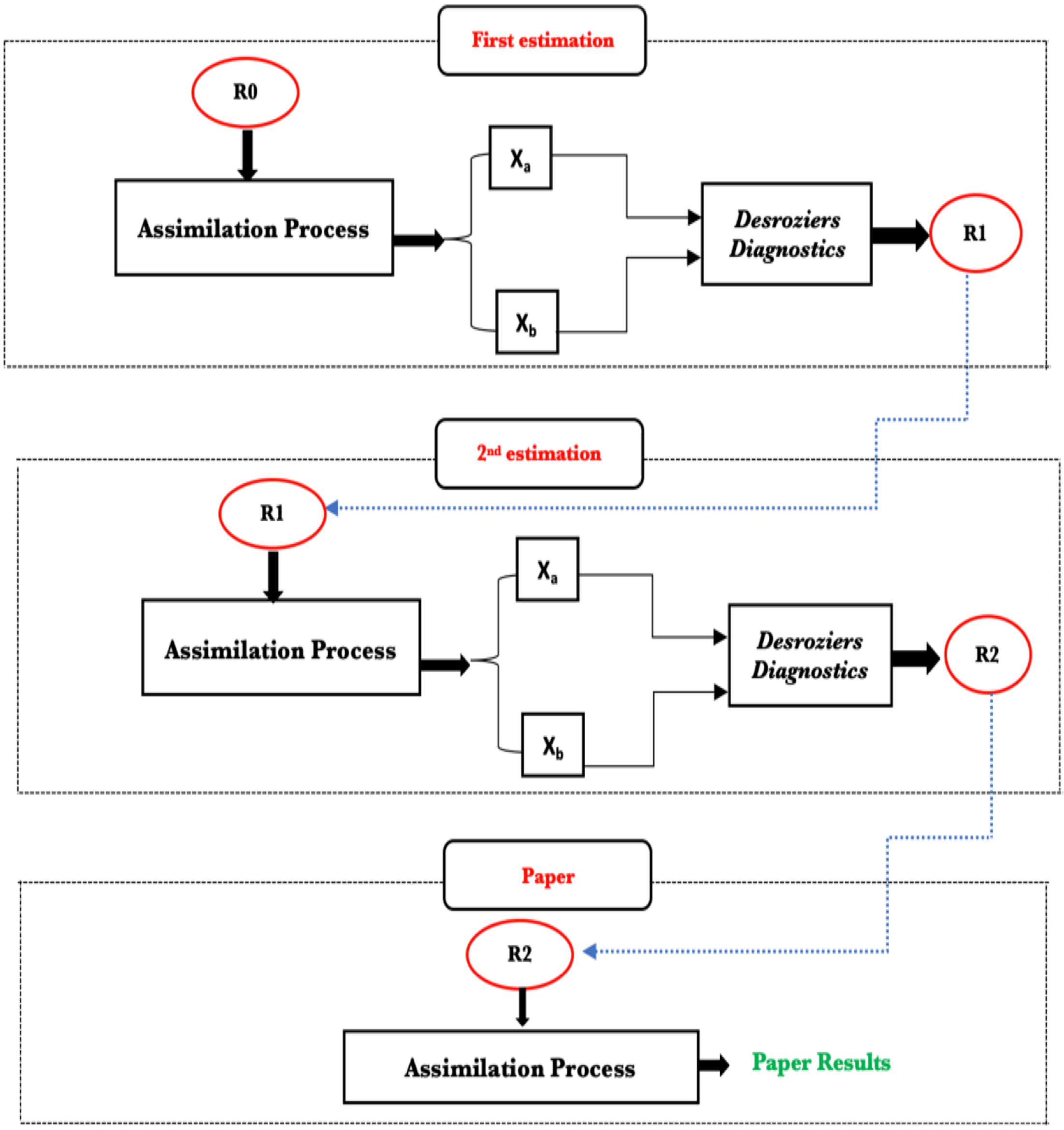


Figure 3_R1 : different estimations discussed in the paper and the one used for the results.

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:

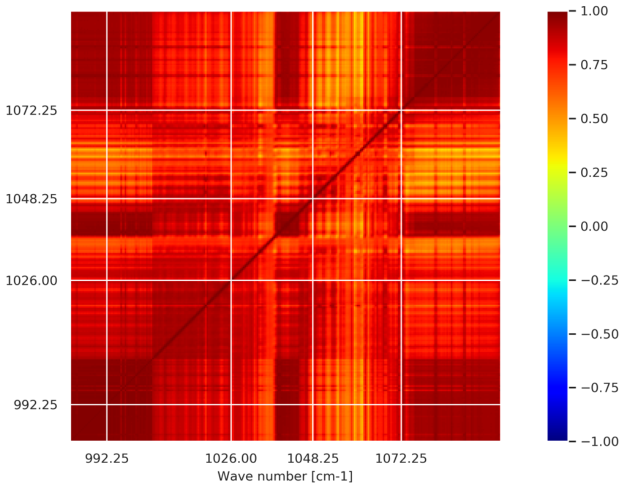


Figure 4_R2: Correlation matrix estimated over the globe (sea and land).

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.

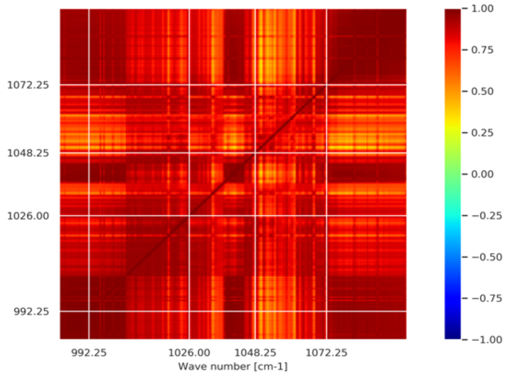


Figure 5b_R1: Correlation matrix estimated over the sea.

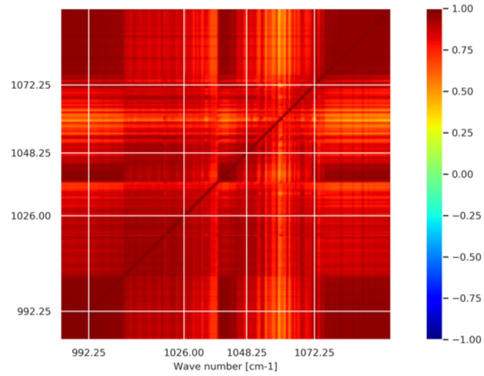


Figure 5c_R1 : Correlation matrix estimated over the land.

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.

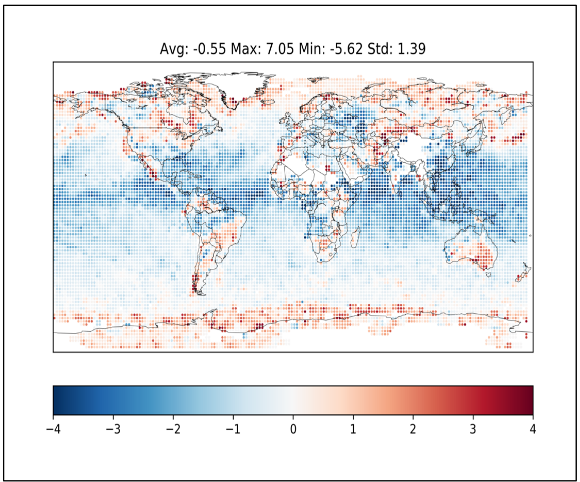


Fig7a_R1. Difference (in °C) between the IFS SST forecast and the analysis of the SST given by RdiagExp (averaged by grid box).

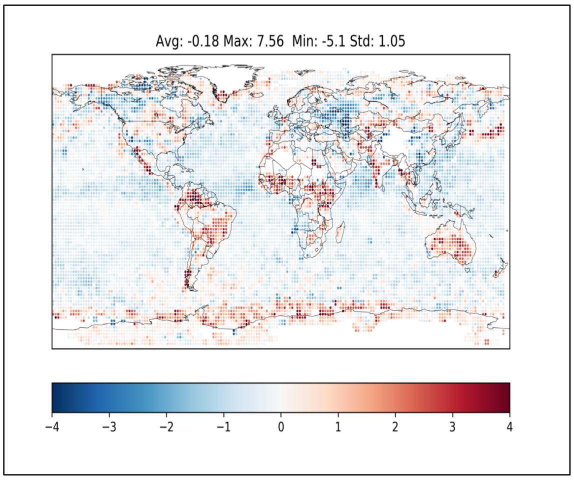


Fig7b_R1. Difference (in °C) between the IFS SST forecast and the analysis of the SST given by RfullExp (average by grid box).

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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