Combined assimilation of IASI and MLS observations to constrain tropospheric and stratospheric ozone in a global chemical transport model: reply to the reviewers

Emanuele Emili[1], Brice Barret[2], Sébastien Massart[3], Eric Le Flochmoen[4], Andrea Piacentini[5], Laaziz El Amraoui[6], Olivier Pannekoucke[7], and Daniel Cariolle[8]

[1,5,7,8]CERFACS, Toulouse, France
[6,7]Météo France, Toulouse, France
[2,4]Laboratoire d'Aérologie, Toulouse, France
[3]ECMWF, Reading, United Kingdom

1 Reply to A.J. Geer

We thank A.J. Geer for the positive evaluation of the manuscript and the constructive comments. The detailed replies to the reviewer's comments follow.

1. The abstract has been revised as follows to incorporate the reviewer's suggestions:

Accurate and temporally resolved fields of free-troposphere ozone are of major importance to quantify the intercontinental transport of pollution and the ozone radiative forcing. We consider a global chemical transport model (MOdèle de Chimie Atmosphérique à Grande Échelle, MOCAGE) in combination with a linear ozone chemistry scheme to examine the impact of assimilating observations from the Microwave Limb Sounder (MLS) and the Infrared Atmospheric Sounding Interferometer (IASI). The assimilation of the two instruments is performed by means of a variational algorithm (4D-VAR) and allows to constrain stratospheric and tropospheric ozone simultaneously. The analysis is first computed for the months of August and November 2008 and validated against ozone-sondes measurements to verify the presence of observations and model biases. Furthermore, a longer analysis of 6 months (July-December 2008) showed that the combined assimilation of MLS and IASI is able to globally reduce the uncertainty (Root Mean Square Error, RMSE) of the modeled ozone columns from 30% to 15% in the Upper-Troposphere/Lower-Stratosphere (UTLS, 70-225 hPa). The assimilation of IASI tropospheric observations (1000-225 hPa columns, TOC) decreases the RMSE of the model from 40% to 20% in the tropics ($30°$S-$30°$N), whereas it is not effective at higher lati-

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tudes. Results are confirmed by a comparison with additional ozone datasets like the Measurements of OZone and wAter vapour by aIrbus in-service airCraft (MOZAIC) data, the Ozone Monitoring Instrument (OMI) total ozone columns and several high-altitude surface measurements. Finally, the analysis is found to be insensitive to the assimilation parameters. We conclude that the combination of a simplified ozone chemistry scheme with frequent satellite observations is a valuable tool for the long-term analysis of stratospheric and free-tropospheric ozone.

2. The nonsensical phrase has been removed. The reader is now addressed to Appendix A (at the end of this document and in the revised manuscript) for a detailed discussion on the impact of the climatological relaxation term on the assimilation. This discussion has been placed in the appendix and not in the main text because it is based on results that are presented later in the manuscript.

3. MLS provides NRT products about 4 hours after the overpass. The IASI SOFRID product that have been used in this study is still in a pre-operational phase, but production within a delay of about 6-12 hours is possible. The information about the NRT availability of satellite data has been added to the satellite data description.

4. We chose the same acronym as in Barret et al. (2011). Since we introduce the total ozone columns only in one figure we decided to keep it, but in all figure legends the words tropospheric column have replaced TOC for sake of clarity. Use of the word TOC in the text has also been generally reduced and replaced with tropospheric column.

5. The argument was effectively not clear and was removed since it does not add useful informations to the manuscript.

6. The sentence has been reformulated as follows:

It was shown that in the upper troposphere and in the lower stratosphere the linear parametrization gives satisfactory results (Cariolle and Teyssedre, 2007) and provides good performances in combination with satellite data assimilation (Geer et al., 2006, 2007).

7. Some of the references have been removed, the ones which employed the 3D-FGAT algorithm to assimilate IASI or MLS data are kept (Massart et al., 2009; El Amraoui et al., 2010; Barré et al., 2012).

8. The text has been updated following the suggestion of the reviewer

9. The aircraft data fully support the ozone-sondes validation, this is the reason why MOZAIC data are ignored after. The text has been changed as follows:

The scores are in agreement with those obtained using sondes data (cf. Fig. 6, at 200 hPa level) and confirm a modest improvement of the correlation and the RMSE for the IASI-MLS

analysis in August and a slight worsening in November. Since the validation with sondes and aircraft data shows a good agreement but sondes have a better global and vertical coverage (cf. Fig. 1), only sondes validation will be shown hereafter.

10. A mention to the method suggested by the reviewer has been added in the text:

    Additional possibilities exist: for example the verification of the MLS+IASI 4D-VAR background against sondes data can be further used to update **B** and recompute a new analysis. However, this method was not tested in the framework of this study.

11. All the vertical profiles plot now contain more labels (as in Fig. 4 of this document)

12. Back-trajectories for the air masses reaching Mauna Loa in August and November 2008 and corresponding to the ozone minima have been computed with the HYSPLIT model (http://ready. arl.noaa.gov/HYSPLIT.php). Most of the trajectories indicate the origin of the air masses from the generally clean Pacific Ocean boundary layer, which might explain the low concentration of measured ozone. For the episodes of 15th August and 21st November, the free simulation (control run) is already able to capture the occurrence of the minima and their temporal duration. This means that those two episodes are mostly related to transport processes. In all cases the local atmospheric circulation is dominated by easterly winds. An analysis of the O_3 field and the horizontal winds at 800 hPa (Fig. 1) confirms that the air masses come from the Equatorial low-ozone belt before the the occurrence of the minima. The end of the episodes is marked by a change of the circulation (Fig. 1, right plots), which brings back ozone-rich air from the Northern latitudes. The overestimation of the measured minima is likely due to the fact that the model is positively biased below 800 hPa over oceans by about 20%, as indicated by the tropics and southern hemisphere sondes validation (Fig. 13 in the manuscript). Furthermore, this also explains why the IASI analysis does not permit to better describe these minima with respect to the control run. Since IASI observations are little sensitive to the planetary boundary layer, the positively biased ozone concentration in this layer is not corrected by the assimilation (cf. Fig. 13 in the manuscript). The episode on 3rd November has a shorter duration (2 days) and it is badder reproduced by the control simulation. The availability of an ozone-sonde measurement on the same day shows that the O_3 has a sharp minimum at about 700 hPa (Fig. 2). Such smaller time and space scale events are difficult to capture with a $2°$ grid resolution model and would probably benefit from the increase of the model spatial resolution.

    We decided to omit the details of this discussion in the manuscript for brevity reasons. The following clarification has however been added to the manuscript:

    In particular, the occurrence of ozone minima ($\sim$20 ppbv) lasting more than 2-3 days in the Mauna Loa time series is mostly due to the transport of low-ozone air masses from the equatorial boundary layer (below 700 hPa). The duration of these episodes is well captured by the
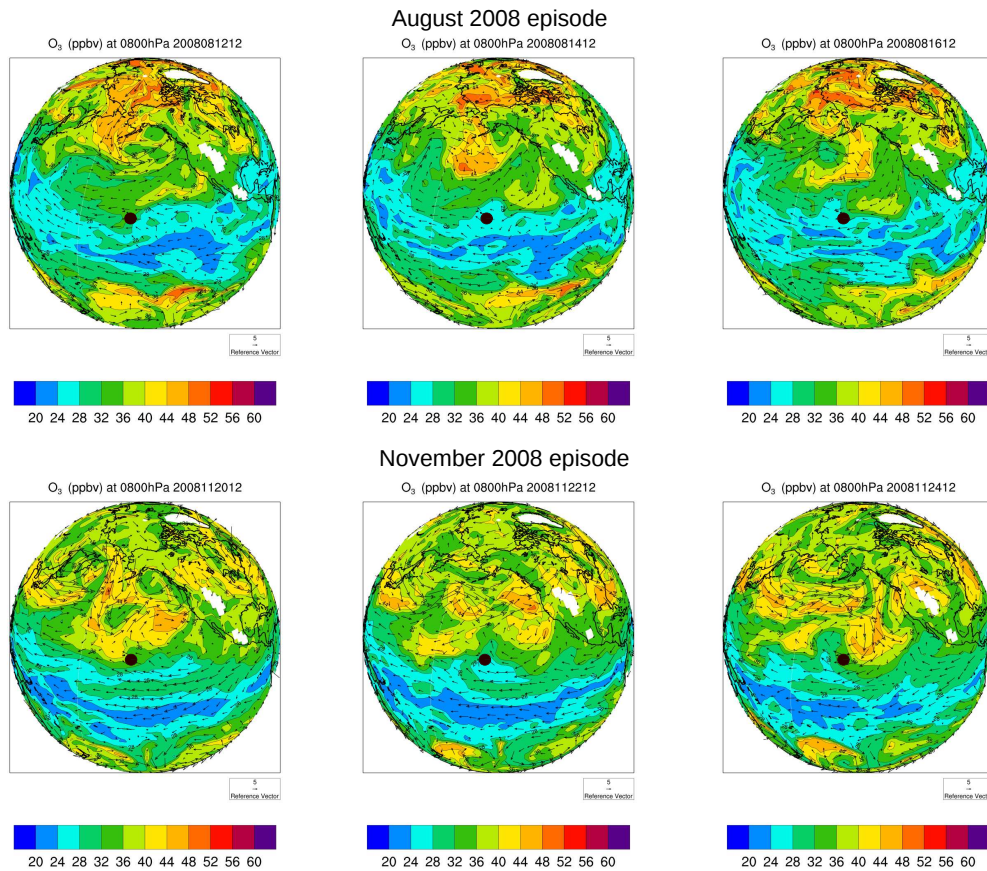
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August 2008 episode

O₃ (ppbv) at 0800hPa 2008081212

O₃ (ppbv) at 0800hPa 2008081412

O₃ (ppbv) at 0800hPa 2008081612

20 24 28 32 36 40 44 48 52 56 60

20 24 28 32 36 40 44 48 52 56 60

20 24 28 32 36 40 44 48 52 56 60

November 2008 episode

O₃ (ppbv) at 0800hPa 2008112012

O₃ (ppbv) at 0800hPa 2008112212

O₃ (ppbv) at 0800hPa 2008112412

20 24 28 32 36 40 44 48 52 56 60

20 24 28 32 36 40 44 48 52 56 60

20 24 28 32 36 40 44 48 52 56 60

Fig. 1. Control run ozone concentration and horizontal wind field at 800 hPa during the two low-ozone episodes at Mauna Loa (19.54 N,155.58 W, indicated with a black circle on the maps, cf Fig. 14 in the manuscript for the time series): top) August episode (13-18/08/2008), bottom) November episode (21-25/11/2008). The reference wind vector is displayed on the bottom of the plots and corresponds to a wind intensity of 5 m/s.

90   control run but their amplitude is not, and IASI low sensitivity to the lower vertical layers does not permit to account for it.

13. All typos and grammar comments have been considered

2   Reply to Referee n. 2

We thank the anonymous referee n. 2 for the constructive review of the manuscript. The detailed
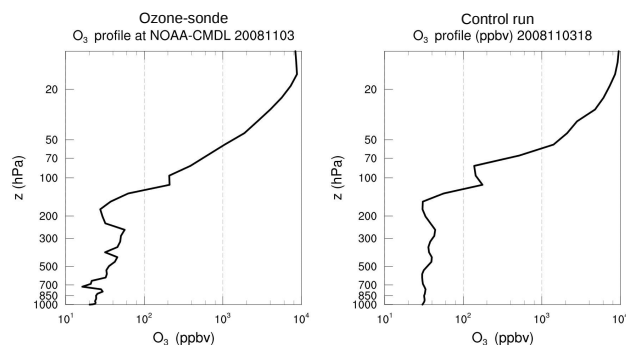95 replies to the reviewer's comments follow.

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Fig. 2. Vertical ozone profile at Mauna Loa (19.54 N,155.58 W) during the low-ozone episode on 3rd November 2008: right) Ozone-sonde measurement at the nearby site of Hilo, Hawaai, at 18 UTC, left) correspondent control run profile

2.1 Abstract

The abstract has been revised and contains now the information about the employed chemical scheme (cf. reply n. 1 to the first referee). The conclusion about the sensitivity of the analysis to the chemistry scheme has been removed. However, a discussion about the impact on the analysis of using a more detailed ozone chemical/physical model is reported here.

To corroborate the conclusion that the CTM complexity does not have a significant role in the global IASI and MLS analysis, we repeated the assimilation experience with a more complex CTM. The chemical scheme used for this purpose is RACMOBUS, which is a combination of the stratospheric scheme REPROBUS (Lefèvre et al., 1994) and the tropospheric scheme RACM (Stockwell et al., 1997). It includes 119 individual species with 89 prognostic variables and 372 chemical reactions. Principal tropospheric dynamical processes are also implemented: turbulent diffusion is calculated with the scheme of Louis (1979) and convective processes are represented following the scheme of Bechtold et al. (2001). Emissions for 2008 are taken from the MACCCity database (Granier et al., 2011) and from the Emission Database for Global Atmospheric Research (EDGAR, http://edgar.jrc.ec.europa.eu) for species not available in the first dataset. The MOCAGE horizontal grid is the same as in the previous simulations as well as the meteorological forcing. However, the vertical grid for the RACMOBUS configuration has a lower number of sigma-hybrid levels (47 levels up to ~10 hPa, instead of 60), thus does not cover the entire stratosphere. Nevertheless, the vertical resolution is slightly improved with regards to the 60 levels configuration (~700 m at the tropopause versus ~800 m previously). The lower number of levels partly balances the increased computational cost of the detailed chemical and physical processes. Since no adjoint of the chemistry/physical modules is still available for this configuration, the 4D-VAR assimilation algorithm is substituted by a 3D-FGAT one, with 8 assimilation windows of 3 hours duration each day. All the other assimila-

tion parameters are kept the same as before. The calculation time required to compute one day of
120 analysis increases from 12 minutes with the 4D-VAR and the linear scheme to approximately 163
minutes with the 3D-FGAT and the RACMOBUS scheme on the same machine.

The 6 months analysis (July-December 2008) presented in the manuscript is repeated with the
detailed chemistry configuration, hereafter named RACMOBUS analysis. The initial condition is
obtained from a spin-up of 1 month starting from a June climatology that considers all the RAC-
125 MOBUS species. A control run without data assimilation is also computed in parallel.

Tropospheric columns (1000-225 hPa) of ozone for August 2008 and November 2008 are com-
pared in Fig. 3, for the control run and the analysis with the RACMOBUS chemistry configuration.
As expected, the inclusion of ozone precursors in the RACMOBUS control run permits to reproduce
the summer ozone maxima over industrialized regions (North America, Europe, Russia and China),
130 the biomass burning and the rain forest VOCs emission patterns in South America and Africa. These
features were not captured by the linear chemistry control run (Fig. 8 of the manuscript). The two
IASI+MLS analyses (with RACMOBUS and the linear ozone chemistry) show instead very similar
geographical patterns and global averages for both periods. IASI observations tend to slightly de-
crease the ozone columns produced by the RACMOBUS simulations (e.g. over Central Europe, US
135 East coast, Russia and China) whereas they increased them with the linear chemistry configuration.

The validation of the RACMOBUS control run and analysis with the ozone-sondes is summarized
in Fig. 4. The linear chemistry analysis is also reported for comparison intents. The figure shows
that the RACMOBUS control run is generally more accurate than the linear chemistry one (Fig. 13
of the manuscript), as we might expect from a detailed chemistry scheme. The South Pole ozone de-
140 pletion is better reproduced, as well as the low-latitudes free-troposphere variability. However, the
RACMOBUS simulation has a positive bias in the tropical and northern latitudes boundary layer,
which might be caused by overestimated precursors emissions or inaccurate physical parametriza-
tion. On the other hand, the two analyses show very similar performances, with the original 4D-VAR
one appearing even slightly superior (by 5-10%) at tropopause altitudes ($\sim$200 hPa). This could be
145 explained by the capability of the 4D-VAR algorithm to better take in account the fast ozone dy-
namics at the tropopause. We can conclude that the choice of the ozone chemistry mechanism is
relatively uninfluential for the purposes of this study.

We prefer not to add this discussion to the study to avoid increasing too much the length of the
manuscript, which is focused on the performances of one CTM and one assimilation configuration.

150 **2.2 Introduction**

1. A standard reference in matter of atmospheric chemistry has been added to the text (Seinfeld
   and Pandis, 1998)

2. The study of Parrington et al. (2008) has been added among the references. The main results
   are now summarized and some additional arguments for exploiting IASI data are given in the
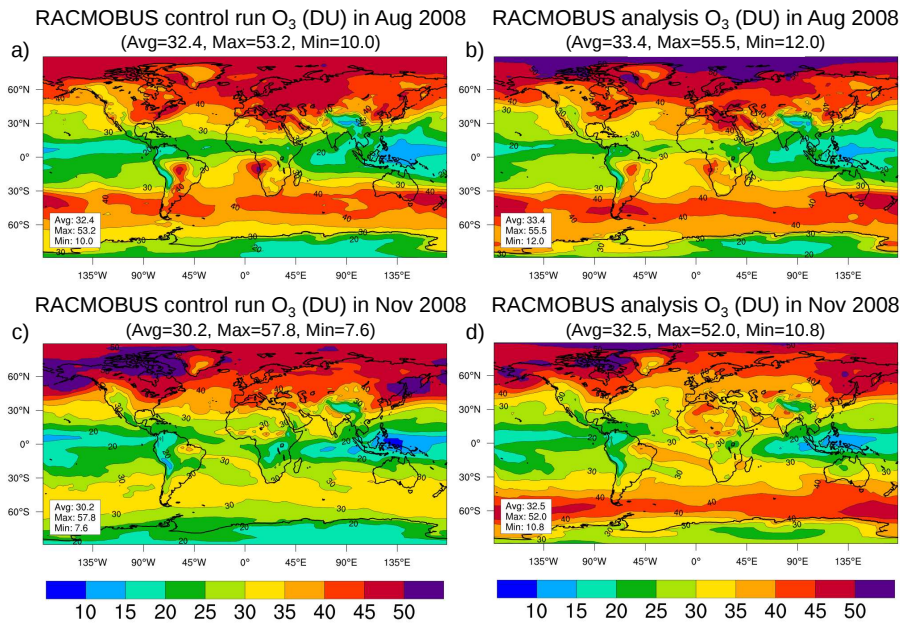
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RACMOBUS control run O$_3$ (DU) in Aug 2008
(Avg=32.4, Max=53.2, Min=10.0)

RACMOBUS analysis O$_3$ (DU) in Aug 2008
(Avg=33.4, Max=55.5, Min=12.0)

RACMOBUS control run O$_3$ (DU) in Nov 2008
(Avg=30.2, Max=57.8, Min=7.6)

RACMOBUS analysis O$_3$ (DU) in Nov 2008
(Avg=32.5, Max=52.0, Min=10.8)

Fig. 3. Average ozone tropospheric columns (1000-225 hPa): **a)** control run with RACMOBUS chemistry for August 2008 **b)** MLS+IASI analysis with RACMOBUS chemistry for August 2008 **c,d)** The same fields for November 2008. Blue/purple end colors represent values that fall outside the color scale.

155   manuscript:

Parrington et al. (2008, 2009) and Miyazaki et al. (2012) assimilated TES data to constrain tropospheric ozone. In most of the cases the bias of the respective models with regards to ozone-sondes data is reduced, although TES data seem to increase biases when the direct model is already unbiased (Fig. 11 in Miyazaki et al. (2012) and Fig. 3 in Parrington et al.

160   (2009)). Few studies explored the assimilation of ozone data from IASI, which is the only sensor sensitive to tropospheric ozone and with a global daily coverage (night and day). IASI increased sampling with respect to TES might improve even more the analysis scores by better constraining the ozone dynamics of the model.

3. The paper of Barré et al. (2013) has been included and the results of the two IASI analysis

165   have been better summarized to put the manuscript into a perspective with similar studies:

Coman et al. (2012) and Barré et al. (2013) assimilated IASI 0-6 km ozone columns in two regional CTMs during a summer month and found improved ozone concentrations with respect to aircraft and surface data, but the limited availability of ozone-sondes data did not allow to draw robust conclusions for the free-troposphere. To our knowledge there is still no study that

170   examined the assimilation of IASI tropospheric ozone columns globally and for long periods.
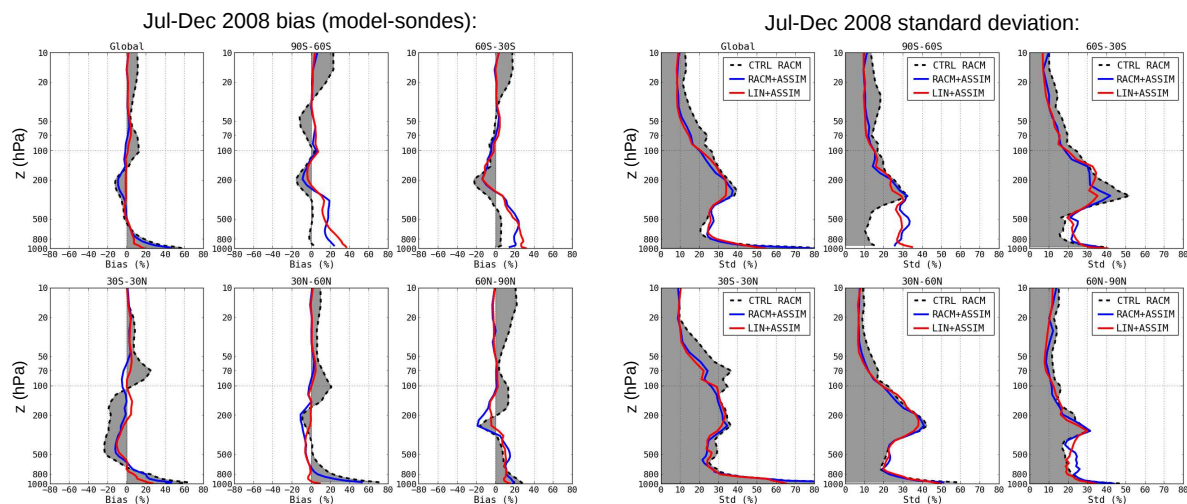
Fig. 4. Global and zonal validation of MLS+IASI analysis versus ozone-sondes profiles for the RACMOBUS simulations (July-December 2008): **left plots)** bias (model-sondes) normalized with the climatology (control run in black, RACMOBUS analysis in blue, linear chemistry analysis in red) **right plots)** standard deviation normalized with the climatology. Positive/negative values in the bias plots mean that the model overestimates/underestimates the ozone-sondes measurements.

2.3 Ozone observations

The amount of pixels removed by the filter have been reported in the text, together with an additional explanation of the reasons to implement it:

The filter removes 25% of IASI retrievals globally, most of them located over ice covered surfaces, mountains or deserts, where the sensitivity of IASI to the tropospheric ozone spectral signature is sensibly decreased (Boynard et al., 2009).

The value of 0.6 has been chosen on the base of the histograms of Fig. 1 in Dufour et al. (2012). Some tests have been done with values of the threshold set to 0.4 or 0.8 but the value of 0.6 gave the best compromise in terms of removal of pixels over difficult surfaces (deserts, ice, snow) and in terms of analysis quality.

2.4 Model description

1. Done

2. The sentence has been corrected and the references revised (cf. reply n. 6 to referee 1). A detailed discussion of the weaknesses of the direct model is better left for the results and discussion section, where it is supported by the various figures.

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2.5 Results and discussion

1. The word chemistry has been substituted by chemical and physical processes, to indicate all the processes which are not described by the linear parametrization.

2. The correspondent vertical length of 1 model grid point is now given in meters inside the brackets:

   ($<$700 m in the troposphere, $\sim$800 m at the tropopause and $<$1.5 km in the stratosphere)

3. The discussion has been extended as follows:

   Ozone underestimation appears also well correlated with regions of high natural VOCs emissions from tropical forests, such as the Amazon and the African rain forests (Guenther et al., 1995).

4. The aircraft data fully support sondes validation, this is the reason why MOZAIC data are ignored subsequently. The text has been made clearer. See reply n. 9 to the first referee.

5. The results of Coman et al. (2012) and Barré et al. (2013) are compared to the outcomes of this study as follows:

   With respect to the studies of Barré et al. (2013) and Coman et al. (2012) on the European domain, we found similar conclusions about the capacity of IASI to reduce the model free-troposphere bias at northern mid-latitudes (30°N-60°N). However, it is found that IASI was not able to improve the model variability (standard deviation) in this region.

6. We think that the main reason for this difficulty is the following: tropospheric ozone concentration has a lower variability at high latitudes than in the tropics or in polluted mid-latitude regions. Even in the case of a very simple tropospheric model, such as the one used in this study, the validation scores are already quite accurate at high latitudes (cf. Fig. 13 in the manuscript). Therefore, very precise observations are required to improve the scores of models. IASI observations do not fulfill this requirement in our opinion, since their sensitivity to the tropospheric ozone signature tend to decrease at high latitudes, due to a reduced surface-atmosphere thermal contrast. This was already somehow explained in the text but the following sentence was added for the sake of clarity:

   However, additional validation studies of IASI products would be required to quantify tropospheric retrieval errors at high latitudes.

7. We agree with the reviewer: MOZAIC data would permit to asses the intra-daily variability of ozone profiles. However, only the airport of Frankfurt, Germany, has several profiles per day during the period of study (2008). To better detect ozone transport events, a 24 hours data coverage is more favorable, and this is the reason why we looked only at in-situ measurements.

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8. The in-situ data have been averaged in time using a moving window of ±6 hours. With this averaging we better highlight the ozone variability signal attributed to synoptic mass transport. We assume in this paper that a 2x2 degree model is able to capture this signal. In fact most of the ozone maxima and minima in the Mauna Loa time series are already reproduced by the control run. On the other hand, the amplitude of those events is mostly missed by the free model and it is better captured by the analysis (see also the reply n. 12 to the first referee for additional details). The two considered sites are located on a barely vegetated mountain in the middle of the Pacific Ocean and on the ice-covered Greenland plateau. Ozone deposition rate is normally much smaller for this surface type than for vegetated continental surfaces (Seinfeld and Pandis, 1998). Therefore, we think that possible local effects due to dry deposition are not among the principal causes of the examined ozone variability.

Appendix A   On the influence of the ozone climatological relaxation

A linear ozone chemistry scheme has been employed in this study (Sec. 3.1 of the manuscript). The main drawback of this scheme is the missing modeling of tropospheric ozone sources, sinks and chemistry. These processes are replaced by a relaxation to a zonal climatology to avoid tropospheric ozone accumulation due to the vertical transport during long simulations. This appendix clarifies the deficiencies of the climatological relaxation within the adopted data assimilation framework. For example, the climatological relaxation counteracts the assimilation increments and might lessen the adjustments produced by observations.

A model simulation has been initialized on 5th July 2008 at 00 UTC using the ozone field calculated from the 6-months analysis (Sec. 4.3.2 of the manuscript). On this date the ozone field has been well constrained by the observations assimilated during the previous days (Fig. 10 of the manuscript) and it represents an initial condition quite distant from the model climatology. Starting from this initial condition, a free model simulation of 24 hours is computed and compared to a second one obtained with the chemistry module deactivated. The latter represents the evolution of a passive tracer. This permits to assess the impact of the ozone chemistry on the temporal evolution of tropospheric columns (1000-225 hPa). The difference between the 2 free simulations after 12 hours is depicted in Fig. 5a. We remark that the ozone partial column is decreased by 0.3 DU with regards to the global average, by a maximum of 1.8 DU at the tropics, where the relaxation term is stronger due to the larger departures from the climatology. These values can be compared with the increments produced by the observations assimilated in the analysis during the same time window (Fig. 5b,c). Absolute increments are spread globally and peak at about 8 DU. This example supports the hypothesis that the chemistry relaxation term plays a relatively minor role, given the global coverage of IASI observations.

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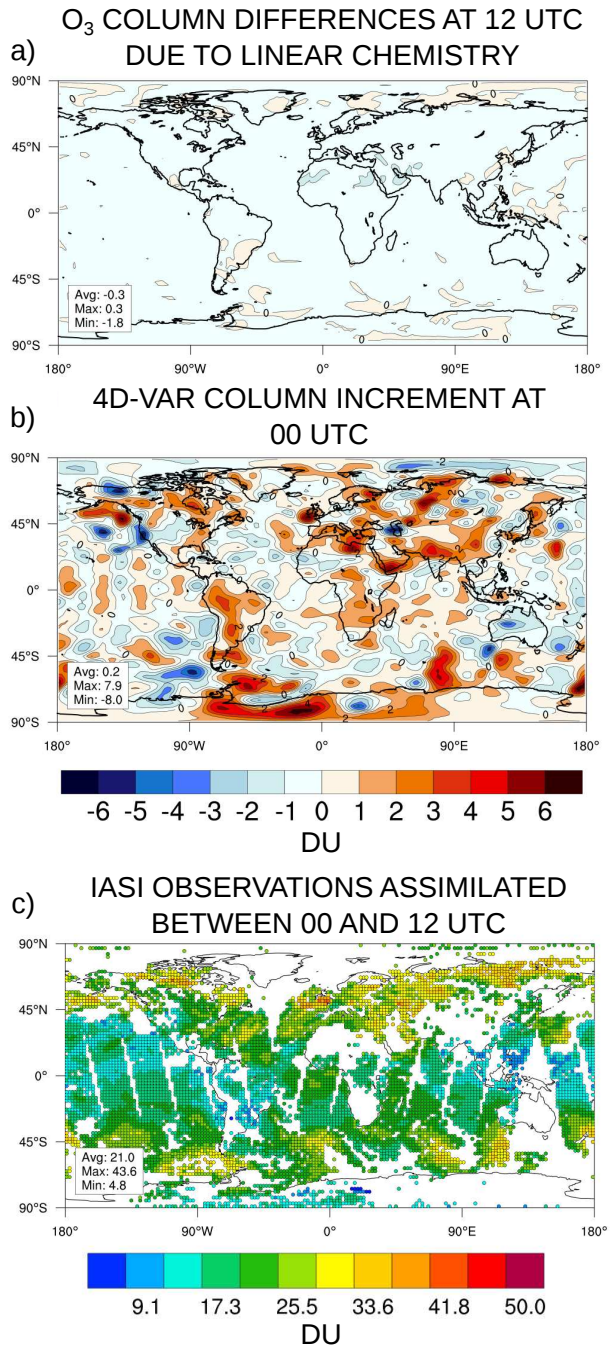
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Fig. 5. Variability of ozone tropospheric columns (1000-225 hPa) during one assimilation window (12h) on 5th July 2008: **a)** difference between a free simulation with linear ozone chemistry and a free simulation with chemistry deactivated (passive tracer) after 12 hours of integration **b)** Increments added at 00 UTC by the assimilation of satellite observations **c)** IASI observations assimilated during the first window (0-12 UTC). IASI values are computed as in Fig. 3,4 of the manuscript. Blue/red end colors represent values that fall outside the color scale.