Response to reviewer #2

We thank reviewer#2 for the insightful and challenging comments on the paper, which instigated a revision of the MOZART-3 model code used in this study. After being granted an three-month extension of the response period, we are happy to present a greatly improved simulation of the ozone hole with IFS-MOZ. All other findings of the original paper remain valid.

Reviewer #2 stated a lack of originality of the manuscript because of an inadequate simulation of the ozone hole without assimilation, in particular by the full scheme of the MOZART-3 model. In a further response to this, we would like to clarify that the comparison of the simulation without assimilation is only one aspect of the paper. The paper presents the performance of the chemistry schemes as part of a data assimilation system. We investigated the impact of the schemes on the analyses and the capability of the schemes to benefit from improved ozone initial conditions in forecasts.

We believe that the realism of the analysed stratospheric ozone profiles obtained by a muliti-sensor approach, the inter-comparison of different chemical schemes and the study of the chemical predictability of the ozone hole are important novelty aspects of the paper.

Nevertheless, we can report that an update of the MOZART-3 model code led to a considerable improvement of the simulation of the ozone hole. The average bias over the ozone hole region was reduced by about 20-35 DU and the extent of the ozone hole area was significantly increased (see Figure 1 of this response). It is therefore not the case anymore that the full chemical scheme is outperformed by the simplified schemes.

We have structured our response to the general comments as follows:

- Model performance – MOZART-3 scheme
- Model performance – simplified scheme
- Chemical Predictability
- Role of MLS
- Response to specific and technical comments

<u>Model performance – MOZART scheme</u>

An update of the MOZART-3 model code to version 3.5.02 lead to an significant improvement of the ozone hole simulation. The most important change with respect to the ozone hole simulations was the correction of an erroneous removal of inorganic chlorine and bromine species in the stratosphere. The model version described in Kinnison et al. (2007) et al. was further updated in the following details: (i) correction of photolysis look-up tables, (ii) local conservation of inorganic chlorine and bromine, (iii) update of rate constants according to JPL06 and (iv) improved simulation of surface area density of NAT and ice PSCs. The MOZART-3 code version 3.5.02 is consistent with the code of the Whole Atmosphere

Community Climate Model (WACCM), version 3.5.48, which was validated in SPARC CCMVal (2010). We will update the model description accordingly.

Figure 1 of this response shows the improvement with respect to the simulation of the ozone hole area below 220 DU. The extent of the ozone hole area was increased from about 40% to about 80% of the area given by the analyses. Although the new simulation still underestimates the ozone hole, start and duration of the ozone hole was correctly predicted. Figure 2 shows monthly averaged profiles from the forecast with MOZART-3 version 3.1.and 3.5. The new version 3.5 shows a more realistic shape of the ozone profile in particular in the period October to December 2008 than the version 3.1, which was used in the original submission. However the shape of the simulated profiles reveal deficiencies, which might be related to the cold bias in the upper stratosphere and a too strong vertical transport in the vortex.

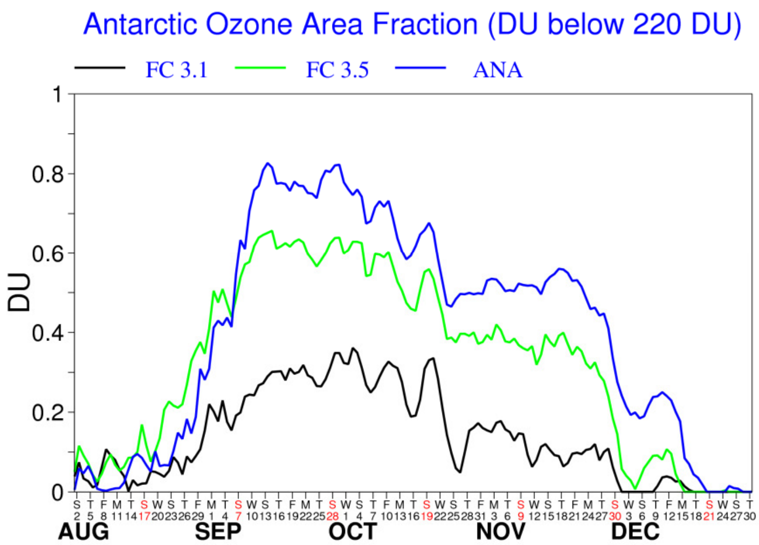

Figure 1 Area fraction below 220 DU in the area 62°S-90°S from the forecasts runs (FC) with IFS-MOZ version 3.1. (used in the original submission), with the improved version 3.5 and from the Analysis (ANA)

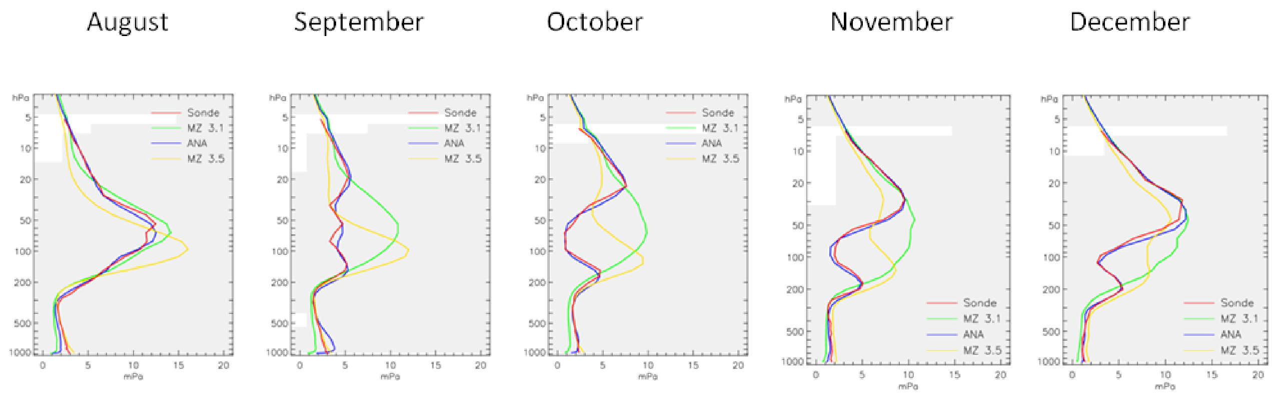

Figure 2 Monthly averaged ozone profiles (partial pressure in mPa) forecast by IFS-MOZART with version 3.1 (used in the original submission) , the new updated version 3.5 (MZ 3.5), the analysis (ANA) and observations (Sonde, red) at Neumayer Station from August to December 2008.

Even with the presented improvements, we acknowledge the need for an improved discussion of the potential reasons for the encountered problems with MOZART-3, which also includes published findings. We will include the following discussion in the revised version of the manuscript.

"Kinnison et al. (2007) report an underestimation of the ozone hole when using meteorological analyses from the ECMWF's operational system and an improved version of the re-analysis system (expid 471). Kinnison et al. (2007) conclude that an overestimation of mixing into the vortex, which they also find in an underestimation of age-of air, is the reason from the underestimation of the ozone hole. Monge-Sanz et al.(2007) and O. Stein (personal communication) reported that the further development of the ECMWF data assimilation system, in particular the transition from 3D-VAR to 4DVAR, greatly improved the realism of the age-of-air simulation The simulation with IFS-MOZ showed that a more realistic ozone hole size and depth can be achieved with the updated version of the IFS meteorological model. There were still deficiencies in the simulation of the vertical profiles, which will be discussed in section 3.4"

In section 3.4, in which the profiles are discussed, we will add the following:

"The ozone depletion in IFS-MOZ is too weak and it occurs at a higher level than the observations suggest. The maximum ozone loss was simulated at a pressure range above 50 hpa whereas the observations showed a stronger ozone decrease between 50 -150 hPa. The identified cold bias above 50 hPa (see Figure 4) could be a reason for an overestimation of the ozone depletion by IFS-MOZ in this region. The same feature, i.e. the overestimated ozone loss above 50 hPa, was also found in the simulation with the linear scheme. The linear scheme responds directly to deviations from the climatological temperature field (see section 2.1.). The lack of sufficient ozone depletion in the lower stratosphere is more difficult to explain but might be caused by an exaggerated downward transport of ozone, which could also be an additional reason for the too strong ozone decrease in the upper stratosphere."

Model performance – linear scheme

We do not consider the performance of the linear scheme and the relaxation to climatology as failure. Both work well within the data assimilation context. The linear scheme, but not the climatological scheme, is able to project the improved initial conditions over a longer period. The simulation of an ozone hole development two weeks too early in the case of 2008, which was a rather late and long-lasting event, meets in our opinion the expectation for such an approach. Our forecast for 2008 seemed to achieve a better agreement with ozone sonde profiles (see Figure 9, middle , panels for August and September) than forecasts for 2001 presented in Cariolle and Teyssedre (2007) as shown in their Figure 13. We will add the following to the manuscript:

“Cariolle and Teyssedre (2007) show that the linear scheme, of which we used version 2a, is able to produce an ozone hole with minimum values of 140 DU, which is about 20 DU higher than the observations from the TOMS instrument for the year 2001”.

Chemical Predictability

Reviewer #2 encouraged us to better explain the concept of the chemical predictability. We will add the following to the manuscript:

“Predictability is understood as the time span over which a satisfactory forecast can be made. The predictability is therefore related to the model performance, which depends on the quality of the model, its initial conditions and boundary data (Kalnay, 2003). The chaotic behaviour of atmospheric dynamics has led to the well known realisation that the predictability of the weather itself is limited (see for an overview Lorenz, 2006, in Palmer and Hagedorn, 2006) and that it varies with respect to the meteorological situation. The impact of the chaotic aspect of the predictability can be quantified with an ensemble of meteorological forecast which use slightly different meteorological initial conditions. These forecasts will diverge more quickly if the predictability is low. The sensitivity of CTMs to initial conditions of the concentrations is strongly controlled by the simulated sink and source terms. Tracers with a long lifetime are longer effected by initial conditions than short-lived tracers. As in the case of the meteorological model , the impact of the initial conditions vanishes but - in contrast to the meteorological case - an ensemble of CTM forecasts will not diverge if the initial concentration of the concentrations were slightly disturbed but they will reach a state controlled by the sources and sinks. “

Within the scope of the paper, we focus on the predictability in respect to the chemical scheme since we mainly use meteorological analyses, and not forecasts, as meteorological fields for the FC15 forecast. We will expand the analysis of the FC15 run by showing the increase in bias of the ozone total columns over Antarctica with increasing forecast length for the different chemistry schemes and additional a tracer without chemistry. The text will be extended as follows:

"As indicated in Figure 7 (bottom), the predictability differed between the time of the ozone hole development and its closure. Therefore the (new) figure shows the development of the bias in total columns over Antarctica separately for the periods 1.8-30.9.2008 and 1.10-31.12.2008. The increase in bias of a the FC15 tracer run without chemistry, which was initialised with ozone analyses as the other FC15 runs, can be considered as reference. It shows the importance of the chemical source and sink at the time scale of the forecast length. The importance of the chemistry during the ozone hole development was obvious since the bias of the tracer run increased to about 35 DU over 15 days. The full chemistry of IFS-MOZ was able to produce nearly un-biased forecasts of the rapid ozone hole development over several days. Negative and positive biases of about +/- 10 DU occurred in the FC15 forecast with IFS and IFS-TM5 respectively after 15 days. During the time of the ozone hole closure, the FC15 tracer forecast had a positive bias of about 15 DU after 15 days. This would mean an overestimation of the ozone transport in to Antarctica, if the chemistry was not important. The linear scheme , however, produced a smaller bias of about 5 DU showing the best predictability of the schemes during the ozone hole closure. The full scheme of IFS-MOZ underestimated the ozone increase at a rate of about one DU per day. The large biases of IFS-TM5 showed the strong limitation of the climatological approach for forecast applications."

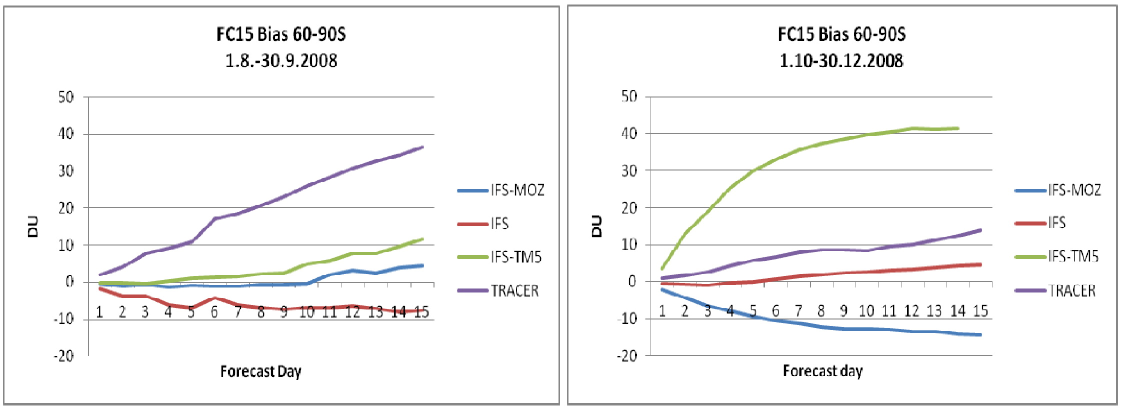

Figure 3 Bias of the total column over 60-90°S of the FC15 runs with IFS-MOZ, IFS, IFS-TM5 and a tracer without chemistry (TRACER) for the period of the ozone hole development (1.8.-30.9, right, average over four FC15 runs) and its closure (1.10-31.12, right, six FC 15 runs). The FC15 runs were initialised with ozone analyses at the forecast start.

<u>Role of MLS</u>

Reviewer #2 does not seem to fully acknowledge the benefit of the multi-instrument assimilation. Reviewer #2 concludes that the analyses are fully controlled by the MLS retrievals. It was one finding of the study that using MLS data greatly improves the analysed profile, but we would argue that combination of high-resolution total column observation (OMI and SCIAMACHY) and the stratospheric profile from MLS leads to the best results . We do not agree with the reviewer's opinion (see specific comments) that the analysis agrees much better with MLS than with OMI and SCIAMACHY. Our study shows (see Figure 3) that the analysis departures, averaged over 2 ° latitude bands, do not exceed 4 DU. OMI is bias-free over the all latitude bands in the considered week at the end of November.

We would like to add the following to the manuscript:

“The assimilated MLS instrument data cover the stratosphere and a further constraint by total column observations is needed to provide information on the troposphere. Several authors (Stajner et al., 2008, Schoeberle et al., 2007 and Ziemke et al., 2006) have used the difference between MLS and OMI to successfully infer tropospheric ozone columns “.

“ The best agreement with the ozone-sonde profiles can be found in November and December, i.e. at a time when the Antarctic a is fully visible for UV-VIS instrument. This indicates a benefit from the assimilation of MLS together with UV-VIS instruments. Also Jackson et al. 2007 report an positive impact by SBUV/2 if assimilated together with MLS. A practical benefit of the multi-sensor approach is that missing or erroneous data will deteriorate the analysis to a smaller extent.”

<u>Specific Comments</u>

p.9175, l.14: forecasting of stratospheric ozone depletion, at least at the time scales studied here, is not important for assessment nor for monitoring purposes.

The emphasize of the sentence was on the forecast of UV radiation, which we believe is an useful application at the considered time scales. To avoid confusion we will rephrase as follows:

“Hence, monitoring of the stratospheric ozone depletion remains important to assess the increase in UV radiation at the surface. “

p.9176, l.25: "... reactive gases such as...": are these 5 species a precise list of the chemical species actually assimilated for GEMS, or is the list actually longer? For the assimilation experiments used here: was ozone the only assimilated species, or were there other species assimilated simultaneously? If yes, what species?

Assimilation experiments have been carried out for all of the mentioned species (see Inness et al., 2009). Only ozone has been assimilated in the experiments presented in the paper.

p.9177, l.7: is the NRT provision of boundary conditions for RAQ forecasts already in place or is it just an important application for the future?

The NRT provision of boundary conditions is in place.

p.9178, l.1: 1-8: this discussion about predictability is quite vague. You should at least provide bibliographical reference(s) for "meteorological predictability" and propose a definition for "chemical predictability" (see general comments).

As mentioned in the section for the general comments, we will expand the discussion of the predictability.

p.9178, l.10-11: the ozone lifetime reported here, and explained in classical textbooks, is based on 2D models with comprehensive chemistry - not on the Chapman cycle alone.

We will rephrase and add a reference for the a 2d model run.

“The lifetime of ozone in the stratosphere varies with height from a couple of hours in the upper part to a couple of month in the lower part as simulated by two-dimensional CTMs (e.g. , Figure 6 in Gray and Coy at al., 1989).”

p.9178, l.13: Eskes et al. (2002) obtain a predictability range of 4 to 5 days - in what altitude range? For the whole total column? If yes, this information is not relevant for present systems which attempt to provide realistic information about the shape of the vertical profile.

Eskes et al. (2002) forecast total columns. We will change the sentence accordingly. We still believe this is relevant since we also discuss the forecast of total columns (e.g. Figure 7)

*p.9178, l.27: This sentence is false in many cases. The initialization of the stratospheric ozone fields is in fact *not* important for CTM runs which last much longer than the longest ozone lifetime encountered in the atmosphere, e.g. 1 year or more.*

We will reformulate as follows:

“.... but also for independent CTM runs, which are shorter than one year.”

p.9179, l.6: While the ozone hole size below 220DU is a classical diagnostic, it would be useful to provide a bibliographical reference where the value of this diagnostic is discussed.

We will include
“A discussion of the usefulness of this frequently used diagnostic is given in Newman et al., (2004)”

p.9180, l.8: Cariolle and Teyssedre (2007) show 3 forms of this parameterization, all failing to deliver sufficient ozone depletion at 100 hPa (their fig.13). It would still be useful to state which form (i.e. what version of the parameterization) is used here.

We will mention that version 2a has been used. Following a suggestion from reviewer #1, we will add that the coefficients c_1 to c_4 vary with time (month), height and latitude. The coefficient c_5 is based on a prescribed chlorine content of 3.31 ppbv.

As pointed out above we do not believe that the linear scheme has failed to deliver sufficient ozone depletion. Further, we find that the agreement in with sondes is better in our application (see Figure 9, middle , panels for August and September) than in the simulation by *Cariolle and Teyssedre (2007)* as shown in their Figure 13.

p.9180, l.16-19: Kinnison et al. (2007) show (their fig.16) that the ozone hole is not reproduced by MOZART-3 when driven by ECMWF fields and discuss the possible causes. It seems completely in the scope of this paper to push this discussion further, or at the very least to recall it (see general comments).

We will extent the discussion. Please see our reply above on the section for the general comments

p.9182, l.15: Aura-MLS retrievals have a sufficiently high vertical resolution to be described as profiles rather than partial columns. The processing of these observations by the assimilation system is actually another question : from table 1, it appears that the profiles are transformed into 16 partial columns prior to assimilation. Why is this done? Could this result in some loss of information about the profile shape?

It is true that the original MLS retrieval is provided as concentration values at pressure levels. We will motive the conversion to partial columns as follows:

"The vertical resolution of MLS in the considered region is about half as fine as the resolution of the assimilating model. The height of the IFS layers varies from 1 km to 2 km between 150 hPa and 1 hPa, whereas the MLS data have a vertical resolution of about 3 km in this pressure range. It was therefore useful to provide the data assimilation system with partial columns to better account for the difference in the vertical resolution between model and observations. The MLS data were converted in partial column without loss of the vertical resolution."

p.9184, l.18-29: As I understand it, analyses departures from the assimilated observations (fig.3) are primarily a verification tool to check that the assimilation system worked correctly. In this study, several instruments are assimilated and the analyses agree much better with one of them (MLS). While this allows to discuss the OMI-MLS and SCIA-MLS biases, the fact remains that OMI and SCIA observations could not be assimilated as well as MLS. The possible causes should be discussed.

Analysis and forecast departures (AmF and OmF) are often used to check if the assimilation system "draws" toward the data. We used analysis departures to show the inter-instrument biases by calculating the differences with respect to the same "reference" field, i.e. the analysis (see Figure 2). As already discussed in the general section, the biases of the OMI and SCIAMACHI data are small and do not exceed 4 DU.

p.9185, l. 12: where does this maximum bias of 3

The maximum bias of 3% was obtained from an alternative version of Figure 2. It shows that the maximum analysis departure occurred at an measurement value of about 120 DU, i.e. 4DU are 3 % of 120 DU.

p.9186, l. 10: Please adapt the CTM description to the topic under study. Wild-fire emissions are completely irrelevant here.

Wild-fire emission may play a minor role for the errors in the troposphere, but we will the adapt the description since this aspect is not discussed in the paper.

p.9187, l.9-13: This kind of quick-look visual check is not acceptable in a refereed journal. If humidity is relevant to your forecasts of the ozone hole, it must be evaluated and discussed in a statistically meaningful way.

We will reformulate as follows.

"MLS retrievals of water vapor can be used to evaluate the stratospheric humidity for latitudes up to 82°S. The observation error varies from 15% at 100hPa to 4% at 1 hPa. (http://mls.jpl.nasa.gov/products/h2o_product.php). At the time of the PSC formation the MLS water vapor retrievals had values of 2.5-3 ppmv within the polar vortex and 4-5 ppmv outside the polar vortex at 56 hPa. PSCs have been simulated within the polar vortex despite an underestimation of humidity by 1 ppmv inside the polar vortex. Outside the polar vortex the water vapor was 0.5 -1 ppb below the MLS observations."

p.9188, l.13-16: The forecast runs present huge biases w.r.t. observations (figure 6). Please mention that this is discussed in the next section.

We will reformulate
" … whereas the biases of the FC runs varied in time, which will be discusses in the next section."

p.9188, l.23 until end of paragraph: this attempt to justify the failure of IFS-MOZART with inadequate wind fields makes no sense, in view of the failure of MOZART itself (see general comments and comments for p.9180).

At this point, the enhanced Brewer-Dobson-Circulation is only mentioned to explain the overestimation of ozone in SH mid-latitudes. This could be a potential reason for the overestimation of the ozone minima in the ozone hole. The FC15 forecast show (see p. 9190 l. 17) that a correction of the overestimation around the vortex do not greatly improve the ozone hole simulation.

Please find our improved discussion in the general section (Model performance – MOZART scheme)

p.9191, l.17: MLS does contain the information necessary about the shape of the ozone profile, including in the ozone depletion altitude range. It has also been shown *that MLS is the biggest contributor to the analyses. So the only issue here is with respect to the tropospheric part of the profile. Unless the pre-processing of Aura-MLS into 16 partial columns led to some loss of important information...*

Please find our improved discussion in the general section (Role of MLS) and out response to the comment on *p.9182, l.15.*

<u>Technical comments</u>

Table 1: Two different datasets were used for MLS - identify the period for each

Only one data set (with a long label being put in two lines) was used. We will correct the error.

Table 2: Column "FC Length" seems to be in hours? But from figure 7, the lengths of FC15 experiments seem to be 15 days? Please clarify

The length of the individual FC15 forecast was 24 hours as stated in Table 2. The FC15 forecast at the first and the sixteenth day of the month was initialized with the ozone analysis. On the other days, the forecast was initialized from the previous forecast.

Figure 3: the two periods must be labelled more clearly

The figure will be improved.

New references

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