

Interactive comment on “Impact of meteorological analyses and chemical data assimilation on modelled long-term changes in stratospheric NO₂” by L. N. Gunn et al.

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Received and published: 19 September 2012

Response to Comments on

"Impact of Meteorological Analyses and Chemical Data Assimilation on Modeled Long-Term Changes in Stratospheric NO₂"

by L. Gunn et al.

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We thank the referees for their comments and suggestions. Here are our responses.

Reply to referee #1

> It is stated in the paper that the inability of the model (without assimilation) to reproduce the NO₂ trend is attributed to the ERA-40 reanalyses. However, I see other sources of model error that are not discussed in the paper and which should be evaluated in comparison to the error introduced by ERA-40. First, uncertainties in the emission rates at the Earth's surface of CH₄, N₂O, CFCs are not discussed throughout the paper. Is this error negligible and is it quantified? Second, transport errors might be attributed to several other sources than the wind fields. For example numerical diffusion of the advection scheme and the implementation of the wind fields in the model play also an important role. By wind implementation, I mean the way ERA-40 reanalyses are degraded to match the model grid as well as the error introduced by the time interpolation from the ERA-40 6-hourly analyses at the model time step.

Of course it is not only the transport that the assimilation is correcting for, however it should be the largest source of error that is being dealt with here.

There is no error associated with emissions in these runs - the model does not use emissions to calculate tracer values. In common with most 'stratospheric' models SLIMCAT uses surface mixing ratios as the boundary conditions. The values used are global means based on observations (from WMO/UNEP Assessments). N₂O is well mixed in the troposphere. Therefore, this constraint to the observed global mean N₂O values provides a realistic lower atmosphere in the model runs. After transport to the stratosphere N₂O decays to yield NO_y.

It is true that transport errors could be attributed to sources other than the reanalyses and one such source could indeed be numerical diffusion of the advection scheme.

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If this was the case the model results should show a high sensitivity to the model resolution, i.e. numerical diffusion will decrease at higher resolution. The SLIMCAT model uses the Prather (M. Prather, 1986) advection scheme which has been shown to have low numerical diffusion. The inclusion of a higher resolution run (2.8 x 2.8 degree) highlights that numerical diffusion is not a significant source of error within SLIMCAT. We will revise the paper so that it is clear that the comparison of low/high resolution runs makes this point.

Another source of error could be the method for implementing the winds in the model. Chipperfield (2006) explains the implementation of the winds and references therein show that the method used in SLIMCAT (processing of ECMWF spectral coefficients directly to model grid) is one of the better methods. Nevertheless, this is a potential source of error in CTMs.

Overall it is clear from this comment that simply stating that the assimilation corrects for transport errors caused by ERA-40 is oversimplified. In the revised copy of the manuscript the authors we will clarify this point and state (as the referee has highlighted) that there are other small sources of error that are also being corrected for.

> Why did you use ERA-40 reanalyses in this paper instead of ERA-interim? The latter is known to have solved most of the shortcomings found in ERA-40 and cover almost completely the period discussed in the paper (ERA-interim starts in 1979 while the period studied in the paper starts in 1977). Moreover, I found the conclusions made in Sect. 3.4 a bit naive as all the community already knows the issues of a CTM driven by ERA-40 (Monge-Sanz et al., 2007; Meijer et al., 2004). However, it would be interesting to make the same study using ERA-interim to see if those wind fields are well suited to drive long CTM runs as those made by Feng et al. (2007).

ERA-40 reanalyses were used in this study because the paper was originally written

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when ERA-interim was not readily available (and certainly not back to 1979). However, the referee makes a valid point for the present day and we will include a run using ERA-interim in the revised manuscript. This will help highlight the differences between the two reanalysis datasets.

Some conclusions in Sect. 3.4 have been stated before in a number of studies, however, the use of chemical data assimilation in this manuscript is unique. We will rewrite this section to make it clear where we are drawing the same conclusions as previous work, but emphasize the method for revealing it is different and the power of data assimilation in this way is very insightful.

> In the paper, all observations are considered without error? The errors should be taken into account in the discussion, especially to assess if the bias between model runs and observations is significant.

In the revised paper we will include all observational errors in the figures (where possible) and in the text.

> Sect. 2 is confusing in some parts (mainly Sect. 2.2) and I would suggest the authors restructure it. Sect. 2.1 should describe the emission rates at the model surface as well as the setup of the aerosols (as this is a part of the model). Then I would include a new section (which should be Sect. 2.2) which describes the assimilation method (and the HALOE data). In the current manuscript, it is not very clear how the assimilation of CH₄ constrains NO_y. Please clarify. Also I did not understand how HCl constrains the other chlorine compounds. This should be clarified. The new Sect. 2.3 (previously 2.2) should present the model experiments as done in the current manuscript but without the description of the assimilation method.

We will consider this and restructure Section 2. We don't need to discuss emissions

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(see above) but will discuss the surface volume mixing ratio and source of aerosol data. We will change Sect. 2.2 as suggested to introduce the assimilation method and describe HALOE.

The assimilation of CH₄ constrains NO_y in the following way: CH₄ has a compact correlation with N₂O and other long-lived stratospheric tracers. The model tracer-tracer correlations (Plumb and Ko, 2002) is used in conjunction with the observed CH₄ to derived new values for the other tracers. This method is described in Chipperfield et al (2002) but we will give more details in this paper. For HCl a further constraint is applied. The above correlations will provide a value for total inorganic chlorine (Cl_y). HCl is part of Cl_y (often a vary large fraction of it). We used the HALOE HCl to overwrite the model HCl but make sure that its value does not exceed the Cl_y derived from the tracer-tracer correlations. We will clarify this.

> The analysis validation (Sect. 3.1) only uses two ATMOS profiles. These are far too few datasets to support the results in later sections. To improve the validation, other datasets must be used and the authors should also consider the comparison method discussed in Geer et al. (2006). Other datasets can come from the other ATMOS mission (as well as using the complete dataset of the November 1994 mission instead of two profiles), UARS MLS (1991-1997 for O₃, H₂O and HNO₃), CRISTA (two short missions in November 1994 and August 1997, O₃, CH₄, N₂O, HNO₃, ClONO₂, N₂O₅, CFCs), MIPAS (2002-2012, O₃, CH₄, N₂O, H₂O, NO₂, HNO₃, N₂O₅, ClONO₂, CFCs) and ozone sondes. Moreover, why not compare model forecasts against HALOE? This is more a verification than a validation but it would be very interesting to see how SLIMCAT NO₂ agrees with HALOE NO₂.

We will include more ATMOS comparisons and also some CRISTA comparisons to obtain more global coverage. However, in the time available it will not be possible to compare with more instruments. As the focus of the paper is on NO₂, the key species

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to validate are the NO_y species. The long-lived species validation is only needed to validate the assimilation and show how it improves the full profile. For the short-lived species comparisons we are only concerned with NO₂, NO, HNO₃ and ClONO₂. Including CRISTA comparisons will give better validation of these species.

> There are several questions raised by figures 4 and 5 which have no answer in the text. First, it would be important to know if the runs that are compared to HALOE are the analyses or the forecasts (it should be the forecasts). Second, although run B agrees better with HALOE than run A, there are still some differences between run B and HALOE. For example, in Fig. 4, CH₄ 35N-60N at 3.2 hPa, there is a good agreement between run B and HALOE until 1995 while later, run B underestimates HALOE by around 0.05 ppmv. Ideally, assimilated data and analyses should agree within the error bars of the data. If this is not the case, this should be explained. I would suggest the authors use the observation operator of the assimilation procedure to interpolate SLIMCAT at the HALOE location during the SLIMCAT run and save these values. This must also be done for run A even if HALOE data are not assimilated. Doing so, runs and data are averaged in the same way which removes the uncertainties that runs and data can be considered in different atmospheric conditions as, for example, if monthly average of HALOE data are compared to monthly average of SLIMCAT. It would also be useful to know what the assimilation time window is. In other words, do you update the model at every model time step when a HALOE profile is found or is this update done every day or month or for another time period? It is clear that the time difference between the observations and the assimilation should be reasonable in order to guarantee the fact that model and data are considered for the same atmospheric conditions. Finally, section 3.2 is much more verification than a validation, as it compares the analyses against the assimilated data. Thus, I would place it before section 3.1.

The assimilation time window is the basic model timestep (e.g. 30 mins). Any HALOE data available in the window is used for assimilation. Yes, the way figures 4 and 5 were

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produced (different sampling of the model and data averages) could lead to differences. We will redo this analysis sampling the SLIMCAT model the same as HALOE. This will clarify any areas of disagreement. We will also rewrite the text to explain the method.

> *The calculation of the differences between the NO₂ observed total columns and the different runs seems to be done in a too simplistic way. As far as I understood, the comparisons given in Table 2 are based on the global means of the total column from the different datasets for the period 1992-2000. Instead, I would fit some parameters of a model to the different datasets. Then, I would verify that the fit is successful, i.e. the model is well chosen. And finally, I would compare the different parameters obtained by the different fits. I am not an expert on trend studies but I presume that the model to fit should include a periodic component and a linear component. A bibliographic research would certainly allow one to find the correct model to fit. Once this is done, it would be easy to detect any offset between the different datasets as well as to compare the value of the trend and the amplitude of the annual variation. Last but not least, I would use the fitted model to display the NO₂ sunset-sunrise ratio in Fig. 8 instead of the data shown, as it would provide clearer information.*

We will investigate this and apply a trend model. We will use one that is available in Leeds and has been used for solar variation studies (Dhomse et al., 2011).

Specific Comments

We will take onboard all of the specific comments and update the manuscript.

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