Atmos. Chem. Phys. Discuss., 10, C9209–C9221, 2010
www.atmos-chem-phys-discuss.net/10/C9209/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Attribution of stratospheric ozone trends to chemistry and transport: a modelling study" *by* G. Kiesewetter et al.

G. Kiesewetter et al.

gregor.kiesewetter@iup.physik.uni-bremen.de

Received and published: 29 October 2010

We thank Anonymous Referee No. 2 for his/her constructive comments to our manuscript. Where appropriate, original referee comments are quoted in *italicized* font.

We have revised the manuscript, trying to include as far as possible the suggestions by five referees. The most important changes in our manuscript are:

- We have conducted a rerun of the model runs analyzed in the study. In this rerun, the temperature dependence of our linearized ozone chemistry scheme (Linoz) is switched on in the upper stratosphere as well. As a consequence, former Fig. 8 has been dropped.
- We have added a model run in which the empirical polar ozone depletion rate is scaled with $EESC^2$, in order to include an upper estimate for the influence of polar chemistry on mid-latitude column ozone trends. In the runs analyzed in the original manuscript, the polar ozone depletion rate was scaled linearly with EESC.
- The trend analysis methodology has been changed. We now apply the method of connected piecewise linear trends described by Reinsel et al. (2002). This eliminates difficulties due to the misalignment of trends at the intersection of the two trend analysis periods, 1979–1999 and 2000–2009.
- The whole modelled TO3 dataset is analysed in one piece now, including overlaps between ERA-40 and ERA-Interim driven periods. While accounting for an offset between the different meteorological reanalysis periods, equal trends are used for regressing the overlap period, thus increasing the robustness of the analysis.
- A regression analysis of TO3 differences between model and observations is used to remove solar cycle and aerosol signals from the observational time series. This has resulted in a new section (now Sect. 4) and an additional figure (now Fig. 6). The modified observational time series, which is better comparable to modelled ozone, is then used in the trend analysis.
- We have included an explicit analysis of changes in column ozone trends. This has resulted in a new figure (now Fig. 9).
- The analysis of profile trends has been extended to the period 2000-2009.

ACPD
10, C9209–C9221, 2010

- We have tried to make the specific findings of this study clearer. In particular, the abstract has been altered, and the conclusions have been completely rewritten. Although our major conclusions do not change, new points have been added that emerge from the revised regression analysis (e.g. discussion of the significance of trend changes).

Replies to comments by Referee 2:

General remarks.

Reviewer 2 criticized the lacking extent of our study, and stated that the conclusions and take-home message were disappointing to him/her. He/she suggested to alter the focus of our study to address the questions

- *The quantification of the contribution of polar ozone loss to mid-latitude trends.*
- *Is there a trend in lower stratospheric tropical ozone?*
- *Is the ozone layer recovering at the poles and at mid-latitudes. Is there support for studies that state that recovery is already detectable?*
- *How much halogen driven chemical ozone loss has occurred before 1980?*

We thank the reviewer for these suggestions on how to improve our manuscript. We have altered our analysis considerably and tried to include as far as possible the answers this study can provide to the questions raised. In detail, we have done the following to address the reviewer's questions:

ACPD
10, C9209–C9221, 2010

- *The quantification of the contribution of polar ozone loss to mid-latitude trends.*

We have tried to clarify this issue stronger, in particular we have quantified this in the abstract and conclusions.

- *Is there a trend in lower stratospheric tropical ozone?*

This is indeed an interesting question. However, the comparison of the modelled ozone profile trends with observations is not fully conclusive, so we feel that we cannot give a definite answer in the current paper. Nevertheless, we have extended the trend analysis to show both 1979-1999 and 2000-2009 profile ozone trends. In 1979-1999, the model does show a negative trend in lower stratospheric tropical ozone, as mentioned in the ACPD manuscript. However, this trend is not sustained in our model during the 2000-2009 period, except for a marginally significant trend in the lowermost model levels (which are actually located in the upper troposphere in the tropics). We have expanded the discussion of this point in the manuscript.

- *Is the ozone layer recovering at the poles and at mid-latitudes. Is there support for studies that state that recovery is already detectable?*

We have altered our analysis method significantly to explicitly address changes in TO3 trends. The whole model time series is now analysed in one piece, applying the method of piecewise linear trends as discussed by Reinsel et al (2002). The results are discussed throughout the revised version of the paper, in particular in what is now

ACPD
10, C9209–C9221, 2010

Sect. 5.2 and Fig. 9. We find a significant change in TO3 trend for Antarctic spring and summer, and NH mid-latitude summer and fall. However, in these atmospheric regions also the model run with constant chemical composition shows a distinct change in trend, and thus the overall change in trend is partly due to changing meteorological conditions.

- *How much halogen driven chemical ozone loss has occurred before 1980?*

While this is certainly an important question, it is not clear whether this study can provide a real contribution to this discussion. For the calculation of polar ozone loss as a function of time, we make the assumption that the empirical loss rate, initially derived from comparison with ozone observations during the Arctic winter 1999/2000, can be scaled with stratospheric chlorine loading. In addition to the calculations presented in our ACPD manuscript where a linear scaling of the polar ozone loss rate with EESC is assumed, we now also include calculations where we assume that the polar ozone loss rate scaled proportional to $EESC^2$. Both assumptions result in Antarctic total ozone variability and trends that agree well with observations, as shown in the new Fig. 3 (update of old Fig.4) and Fig. 7 (update of old Fig. 5). Fig. 11 (update of old Fig. 10) quantifies the contribution of polar ozone loss to the overall trends, including Antarctic spring. While contributions of changing polar chemistry to total ozone trends seem reasonable, the absolute magnitude of polar ozone loss is challenging to quantify, as considerable differences in absolute polar TO3 losses are encountered between different meteorological datasets. Thus we do not want to make quantitative statements on the amount of polar ozone depletion before 1980 here, as we feel that our simple approach used here can not give reliable answers to this question. Rather we propose to further investigate this question using comprehensive

ACPD
10, C9209–C9221, 2010

full chemistry model calculations. A note to the text has been added.

Detailed comments.

The onset of recovery (page 17493, l. 7).

We have now modified the calculation of trends by using a piecewise linear regression model, which allows us to make statements about the significance of trend changes. We have included a discussion of trend changes and included a figure (new Fig. 9) that shows the significance of the total ozone trend changes for model and observations. We find that while the total ozone trends themselves are not statistically significant over the 2000-2009 period, changes in trends are significant at northern and southern hemisphere mid-latitudes and for Antarctic spring. However, changes in meteorology have contributed roughly half to the trend changes.

A deficit of the study is that it neglects both the influence of the solar cycle and of volcanic eruptions. While the former point results only in a modulation of ozone on a 11-year time scale the latter point is problematic, as sulfate aerosol impacts the ODS driven chemistry that the paper focuses on (e.g., Portmann et al., 1996; Solomon et al., 1996). This aspect should be discussed in more depth in the paper than in the present version.

We have addressed this issue by including a regression of TO3 offsets between model and observations, and constructing an observational time series with solar and aerosol signals removed, which is then better comparable to modelled TO3. This has resulted in a new section (Sect. 4) and an additional figure (Fig. 6).

ACPD

10, C9209–C9221, 2010

The temperature dependence of gas-phase chemistry in the upper stratosphere. [...] Just ignoring the temperature dependence of upper stratospheric ozone is not a solution.

While the upper stratosphere is not of key importance to column ozone trends analysed in this study, we agree that ignoring the temperature dependence in the upper stratosphere altogether is unsatisfactory to the reader, even though observed profile trends are matched better without temperature dependence. We have performed a rerun of the model runs analysed in this study. In the updated version, the temperature dependence is switched on in all but the two uppermost model levels (in these, the high temperature dependence of the Linoz scheme combined with unrealistic temperatures in the meteorological datasets lead to unrealistic ozone concentrations or even model crashes at certain dates). TO3 trends are almost unchanged in this version.

It is stated in the paper that a "significant part of the decadal-scale deviations between modeled and measured TO3 in Fig. 1 appears to be related to the 11 yr solar cycle, which is not directly accounted for in our CTM". However, e.g. using multiple linear regression models and other time-series analysis tools (e.g., Fioletov, 2009; Mäder et al., 2010), it is possible to remove the 11-year solar cycle from the time series. Why is it not attempted here to create such a 'solar cycle free' observational time series for better comparison with the model results?

We thank the referee for this suggestion. We have followed the referee's suggestion and included a multiple linear regression analysis to remove solar cycle and aerosol signals from the observational time series. We use the solar flux and aerosol optical depth to explain the differences between modelled and observed TO3 and then sub-

ACPD

10, C9209–C9221, 2010

tract the corresponding signals from the observational time series. This time series is now better comparable to modelled TO3, and is used in the piecewise linear trend regression.

Column ozone trends: I suggest [...] employing EESC as an explanatory variable.

While there are good arguments in favour of an EESC regression to ozone observations, we have decided to apply the method of (connected) piecewise linear trends as suggested by Anonymous Referee 3 and described by Reinsel et al. (2002). This regression method suits best the model approach taken in this study, as the gas phase (Linoz) ozone chemistry is forced with "piecewise linear" source gas concentrations between 1978-1999 and 2000-2009, whereas polar chemistry is scaled directly to an EESC curve that peaks in 2001. Moreover, the piecewise linear trend model allows us to apply one and the same regression model to all TO3 time series (and their differences) in order to distil contributions from different processes, while there is no reason to apply an EESC fit e.g. to a model run with constant chemical conditions (cc), which is needed to derive the meteorology-induced trend.

Tropical ozone trends.

As stated above, we have added a few sentences to address this issue, and included profile trends for the trend-change period 2000-2009.

Polar ozone trends: Visible in Fig. 4 is a substantial low bias in SH column ozone. This low bias is in contrast with the authors suspecting that they are underestimating the period over which chemical ozone loss occurs in their simplified chemistry scheme.

We thank the referee to point to this issue. Unfortunately, the bias in Antarctic TO3

ACPD

10, C9209–C9221, 2010

shown in Fig. 3 (now Fig. 4) was due to a technical mistake: a wrong climatology (using March instead of October) was used when adding the climatological ozone column below 330K. This is now corrected (new Fig. 4), resulting in a much improved agreement between modelled and observed column ozone south of 63 degrees.

Polar ozone trends: In Fig. 4, the average over the polar region is taken from a latitude of 65°; I recommend using 63° as is conventional.

We have followed the reviewer's suggestion (the concerned figure is now Fig. 3).

Polar ozone loss: I recommend considering to better quantify the chemical polar ozone loss in the model, e.g. by implementing a passive ozone tracer.

It seems that there has been a misunderstanding here. We do not attempt to quantify polar ozone losses in Fig. 3 (formerly Fig. 4) but rather polar ozone evolution. Polar ozone losses are defined in our study as offsets between a model run with polar chemistry and a model run without polar chemistry. We have tried to clarify this in the manuscript. Losses are quantified in Fig. 4 (formerly Fig. 9). These losses are not exactly the same as a passive tracer would show, since gas phase chemistry is not completely negligible in polar winter and spring (see $tc - cc$ curve in the trend attribution plot, Fig. 11, formerly Fig. 10). Changing the method of quantification of polar ozone losses (e.g. averaging poleward of 63equivalent latitude) does not alter the numbers as much as switching the meteorological reanalyses. Thus we conclude that the exact amount of polar ozone loss in our CTM is prone to uncertainties, and should only be quantified approximately as done in the manuscript.

ACPD
10, C9209–C9221, 2010

It is stated that substantial polar ozone loss is present in the model before 1980. But this point is neither more precisely quantified (is it more that 30%?) nor discussed in detail. But of course this is an important point.

See our reply above. There is no doubt about the importance of this scientific question, but we cannot give reliable and robust values with the simple parametrisation employed here. Both tt and tT model runs show substantial polar ozone losses before 1980 (more than 30% of average late-1990 values), but the polar chemistry scheme employed does not seem suitable to address this delicate question with due reliability. It would be interesting to address this issue specifically in a future study with the CTM run in full chemistry mode, including a detailed polar chemistry scheme.

Minor issues.

The presentation and the wording throughout the paper is often somewhat unclear. For example: Linoz-mode and polarchem mode (p.17497). Theta levels (p.17503). Jargon like that should be avoided throughout the paper. Further there are some errors in the English, e.g., tabled should be tabulated.

We have tried to eliminate unclear wording and spelling/grammar errors.

page 17496, l. 16: What is the condition for the onset of ozone destruction?

We have added a sentence in the paper to clarify this issue. From average HNO_3 and H_2O concentrations in the lower stratosphere, we calculate the onset temperature T_{NAT} for Nitric Acid Trihydrate (NAT) formation. In every grid cell that has a temper-

ACPD

10, C9209–C9221, 2010

ature $T < T_{NAT}$ and a maximum solar zenith angle of $92.5°$, ozone is destroyed at a defined rate which has been tuned to match Arctic ozone depletion in the winter of 1999/2000.

page 17498, l. 25-28: The offset gets worse when switching from ERA40 to EI. Isn't that worrying as EI should be the superior product?

The fact that switching meteorological datasets can introduce significant ozone changes in a CTM is well known. Linoz tables use the temperature climatology of Nagatani and Rosenfield (1993), which is obviously better comparable to the ERA-40 fields. While ozone offsets between different reanalysis periods are certainly annoying, they do not affect our trend attribution, as the regression analysis treats each period separately while allowing for an offset.

p. 17501, l. 29: It is not surprising that there are small differences here – EESC does not change much of this time period, so the differences must be small. The analysis should go beyond this point.

It is correct that only small effects are expected from small EESC changes. However, the aim of this study is to quantify the relevance of EESC changes to modelled TO3 trends (and changes in trends), which are in good agreement to what observations show. Mentioning the small differences between tt and cc trends at this stage is an anticipation of Sect. 6, where the different contributions are analysed in greater detail. We believe that the analysis presented in Sect. 5.2 (formerly Sect. 4) has profited from the modification of the regression model to the Reinsel et al. (2002) approach, which allows us to derive a statement about changes in trends and their significance.

ACPD
10, C9209–C9221, 2010

p. 17503, l. 15-18: To me this is bad news. A well known driver of upper stratospheric ozone trends is neglected in the model and the model represents the observations. I would suspect then that the model will no longer represent the observations if the observed temperature trend is taken into account.

As stated above, we have now used a version of the gas phase chemistry that accounts for temperature dependence in the upper stratosphere as well. Some of the difficulties mentioned in the ACPD manuscript (conflicting trends between different meteorological reanalyses) are resolved by the use of the full CTM output, which forces equal trends on overlapping time periods. Switching on the temperature dependence slightly decreased the agreement with observed upper stratospheric trends, but the overall picture does not change much.

We thank the reviewer for several helpful suggestions on how to improve the wording and style of writing.

References.

Nagatani, R. M., and J. E. Rosenfield, Temperature, net heating and circulation, in The Atmospheric Effects of Stratospheric Aircraft: Report of the 1992 Models and Measurements Workshop, NASA Ref. Publ. 1292, edited by M. J. Prather and E. E. Remsberg, pp. A1 – A47, 1993.

Newchurch, M. J., Yang, E.-S., Cunnold, D. M., Reinsel, G. C., Zawodny, J. M., and

ACPD

10, C9209–C9221, 2010

Russell III, J. M.: Evidence for slowdown in stratospheric ozone loss: First stage of ozone recovery, J. Geophys. Res., 108, 4507, doi:10.1029/2003JD003471, 2003.

Reinsel, G. C., Weatherhead, E. C., Tiao, G. C., Miller, A. J., Nagatani, R. M., Wuebbles, D. J., and Flynn, L. E.: On detection of turnaround and recovery in trend for ozone, J. Geophys. Res., 107, 4078, doi:10.1029/2001JD000500, 2002.

Yang, E.-S., Cunnold, D. M., Newchurch, M. J., Salawitch, R. J., McCormick, M. P., Russell III, J. M., Zawodny, J. M., and Oltmans, S. J.: First stage of Antarctic ozone recovery, J. Geophys.Res., 113, D20308, doi:10.1029/2007JD009675, 2008.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 17491, 2010.