

## ***Interactive comment on “Dynamically controlled ozone decline in the tropical mid-stratosphere observed by SCIAMACHY” by Evgenia Galytska et al.***

### **Anonymous Referee #1**

Received and published: 17 September 2018

The authors thank the Referee for his/her thorough reviewing of the manuscript. We address the Referee's criticisms and highlight our proposed improvements below one-by-one in blue. We use the following notation: **P1 L10** means Page 1, Line 10.

This manuscript addresses the trend in N<sub>2</sub>O, and the resulting trend in O<sub>3</sub>, which has been observed in the tropical mid-stratosphere (30-35 km) on decadal scales by several instruments. The overall SCIAMACHY measurements included here also show this trend in O<sub>3</sub>, and the trend in NO<sub>2</sub> which one expects from the dynamical changes which drive the N<sub>2</sub>O trend.

The significant contribution that this manuscript makes, is to show that, according to TOMCAT simulations, there is (from 2004-2012) an increase in Age-of-Air (AoA) in the tropical mid-stratosphere (30-35 km) during some seasons, and a decrease in AoA during others. This result seems plausible, and offers the interesting possibility of changing N<sub>2</sub>O (and hence O<sub>3</sub>) in this region, while perhaps not changing AoA as much as might otherwise be expected.

While this is quite interesting, the authors have somewhat oversold the conclusion. They can conclude from their model that there is “no statistically significant trend in AoA”, but they cannot say that there is “no change in AoA” (in fact, there is a small overall increase in AoA in their model results).

The Referee is correct in the assertion that the absence of significance in annual mean AoA change does not mean the absence of changes in annual mean O<sub>3</sub>. However, we discovered that seasonal changes in AoA are significant and they lead to the non-linearity of the physical-chemical mechanisms controlling the O<sub>3</sub> amount and distribution.

To address this issue we improved the following formulations in the revised manuscript:

- **P8 L7** we replaced ‘The significance of observed changes...’ with ‘The statistical significance of observed changes...’
- **P14 L1** we replaced ‘The absence of AoA changes...’ with ‘The absence of statistically significant AoA changes...’.

However, we disagree with the Referee that our model results show a small overall increase in AoA. In the area of tropical mid-stratosphere, defined in the manuscript on **P3 L5-6** (10°S-10°N, 30-35 km altitude) the negative AoA changes are statistically insignificant (see Fig. 8b). The small region at ~30-32 km altitude and ~10°S in this box exhibit statistically significant negative changes.

While I have no reason to doubt the model results, their explanations for why the seasonal variation in AoA causes N<sub>2</sub>O and AoA change differently do not provide any useful insight. It is, of course, highly desirable to have a better understanding of the N<sub>2</sub>O and AoA relationship, but unless the explanations can be greatly improved I would recommend dropping these from the manuscript.

To address the issue we have rewritten the explanation of the N<sub>2</sub>O-AoA non-linearity. Please, see below our improvements in the 'More detailed comments' and/or **P19 L19-33**.

I also have some serious concerns with the presentation of the SCIAMACHY measurements in the manuscript. The authors need to make very clear to the reader that, **contrary to the model, they have not found any SCIAMACHY data which shows statistically significant increase in SCIAMACHY O<sub>3</sub>, or a decrease in NO<sub>2</sub>, during any particular month or season**. It is certainly not appropriate that the measurements during the months when the model says that an increase in O<sub>3</sub> or a decrease in NO<sub>2</sub> should occur, and which shows no significant measurement trend, are relegated to the supplement, while at the same time the data during months when the opposite trends occur and the model and measurement trends agree (at least in sign and significance) are shown alongside the model in the main text.

The Referee's criticism implies that we have inadequately explained the mechanism which we think explains the behaviour. To address the issues, we have improved the presentation of SCIAMACHY measurements in the manuscript, Specifically, we added SCIAMACHY NO<sub>2</sub> and O<sub>3</sub> data, which showed insignificant gradients/changes, to Fig. 12c,d and we depicted statistically significant (2-sigma) changes as solid lines, and otherwise as dashed lines.

We also rewrote the explanations related to Fig. 12:

- We mention that SCIAMACHY measurements do not yield statistically significant gradients for the time series of Januaries and Februaries in **P19 L2-3**: 'SCIAMACHY measurements show statistically insignificant changes of NO<sub>2</sub> and O<sub>3</sub> during Januaries and Februaries (Fig. 12c,d, Supplements Fig. S4)'.
- We also added that contrary to model simulations, SCIAMACHY measurements do not show a NO<sub>2</sub> decrease and an O<sub>3</sub> increase when analysing changes for any particular calendar month (**P19 L3-5**): "Contrary to the TOMCAT simulations, SCIAMACHY measurements do not show a statistically significant NO<sub>2</sub> decrease and O<sub>3</sub> increase when analysing changes for any particular calendar month'.
- We also discuss possible reasons for the model-measurements differences (Fig. 12c,d) on **P19 L5-11**: "From September to February, the gradient of O<sub>3</sub> time series increases, becoming more positive for both SCIAMACHY and TOMCAT data, resulting for February in small, statistically insignificant negative gradients for SCIAMACHY observations and small but statistically significant positive gradients for TOMCAT. Similarly for NO<sub>2</sub> mixing ratios, from September to February the gradients decrease i.e. they become more positive for both, SCIAMACHY and TOMCAT results. The SCIAMACHY data show larger errors on gradients of the time series for individual months, than those of the TOMCAT model. This results from the stronger oscillating structure in the SCIAMACHY time series. The reasons for the observed oscillations and their strength are not yet unambiguously identified and are under investigation."

More detailed comments (some of which repeat points from above):

Page 6 line 19 – “Global coverage of SCIAMACHY limb measurements was obtained within 6 days at the equator and less elsewhere.” It’s not clear to me what this means. Perhaps the authors are requiring some maximum distance between measurements. Unless the authors wish to provide a clear definition I would recommend dropping this sentence.

We simplified the sentence on **P6 L19-20** in the revised manuscript as follows: ‘For the SCIAMACHY limb measurements, the global coverage was obtained within 6 days.’

Page 6 line 24 – “the errors of single measurements are mostly normally distributed and no additional issues with outliers have been reported.” I think this means that there was no need to remove outliers, but if this is the case please say this more clearly. If this is not the case then please rewrite the sentence to better explain what is meant.

We have reworked the sentence and added the reference to Gebhardt et al. (2014) on **P6 L24-26** as follows: ‘We calculate zonal monthly mean O<sub>3</sub> and NO<sub>2</sub> values as arithmetic means as according to Gebhardt et al. (2014) ‘the errors of single measurements are expected to be normally distributed and no issue with outliers is known’.

Page 6 line 25 – “Consequently, we assumed that the random errors of zonal monthly means could be neglected.” Without knowing at this point how you are using the data it is hard to know whether this is reasonable or not. I would drop this sentence from here and perhaps make the point.

We have withdrawn the sentence. Thank you.

Page 7 – “In the latitudes between 50-60N and within altitude range 15-26 km we applied cumulative eddy heat flux instead of harmonic fit terms. We used ERA-Interim eddy heat flux at 50 hPa integrated from 45N to 75N with the time lag of 2 months.” I am not acquainted with this method. Do other groups do this? Is there a reference? If not, please give some explanation/justification.

This method was previously applied by Gebhardt et al. (2014). We improved the sentence by adding the reference to the method on **P7 L25-29** as follows: ‘At latitudes between 50-60° N and in the altitude range 15-26 km the cumulative eddy heat flux replaced the harmonic fit terms, similar to Gebhardt et al. (2014). The eddy heat flux was used as a proxy for the transport of stratospheric species due to variation in planetary wave forcing (Dhomse et al., 2006; Weber et al., 2011). Here, we used ERA-Interim eddy heat flux at 50 hPa integrated from 45° N to 75° N with a time lag of 2 months’.

Page 13 line 21 –“The absence of AOA changes in the considered region . . .” This is a fundamental conclusion of the paper, but it represents an unjustified conclusion from the statistics. One cannot conclude from the absence of statistical significance that “there is no change in AOA”. One can only conclude that “there is no statistically significant trend”.

We agree with the Referee and we reworked the sentences on **P14 L1-2** as follows: ‘...and according to Fig. 8b there are no statistically significant changes in AoA in the same region. The absence of statistically significant AoA changes here is on the one hand in agreement with ... ’

Figure 9 is particularly interesting.  
Thank you.

Figure 12 – “There are no significant changes in SCIAMACHY measurements taken in February (see Supplements Fig. S4), therefore they are excluded from the figure.” One can’t simply include the SCIAMACHY measurements for a particular month per year when they fit the model, and then ignore them when they don’t. The SCIAMACHY NO<sub>2</sub> results as shown in the supplement are almost significant at the 2-sigma level (they are certainly significant at 1-sigma) and are in the opposite direction of what the model shows. The easiest solution would be for the authors to conclude that the SCIAMACHY measurements, when plotted as one month per year, simply aren’t up to this, and therefore need to be dropped from this figure entirely. The SCIAMACHY results as shown in Figure 2 and 5 certainly do demonstrate the value of this measurements when they are not subsampled as in Figure 12. To address the criticism, we added the SCIAMACHY data for NO<sub>2</sub> and O<sub>3</sub>, which showed insignificant gradients, to Fig. 12c,d (as mentioned above). We plotted statistically significant (2-sigma) linear changes as solid lines and insignificant changes as dashed lines. We also rewrote the explanations related to Fig. 12:

- We mention that SCIAMACHY measurements do not yield statistically significant gradients for the time series of Januaries and Februaries in **P19 L2-3**: ‘SCIAMACHY measurements show statistically insignificant changes of NO<sub>2</sub> and O<sub>3</sub> during Januaries and Februaries (Fig. 12c,d, Supplements Fig. S4)’.
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- We also discuss possible reasons for the model-measurements differences (Fig. 12c,d) on **P19 L5-11**: “From September to February, the gradient of O<sub>3</sub> time series increases, becoming more positive for both SCIAMACHY and TOMCAT data, resulting for February in small, statistically insignificant negative gradients for SCIAMACHY observations and small but statistically significant positive gradients for TOMCAT. Similarly for NO<sub>2</sub> mixing ratios, from September to February the gradients decrease i.e. they become more positive for both, SCIAMACHY and TOMCAT results. The SCIAMACHY data show larger errors on gradients of the time series for individual months, than those of the TOMCAT model. This results from the stronger oscillating structure in the SCIAMACHY time series. The reasons for the observed oscillations and their strength are not yet unambiguously identified and are under investigation.”

Page 19 line 13- This paragraph purports to explain the absence of change in AoA. While it is certainly possible that one could have a change in N<sub>2</sub>O and not a change in AoA, this point has not been proven. At the same time, the explanation seems to be simply a complicated statement of the fact that changes in N<sub>2</sub>O are governed by changes in upwelling speed, which obviously couple to AoA. Unless the authors can offer some additional insight here I would recommend dropping this paragraph.

We reworked the explanation of N<sub>2</sub>O-AoA non-linear relation on **P19 L19-33 as follows**:

'The negative AoA gradients for the 2004-2012 period during the boreal winter months (January and February) and positive AoA gradients during the boreal autumn months (September and October) cancel, i.e. there is no statistically significant linear change/gradient in the annual mean AoA (Fig. 8b). In contrast, the monthly gradients over the same periods for the chemical species  $N_2O$ ,  $NO_2$  and, as a result of the  $NO_x$  ozone catalytic destruction cycle,  $O_3$  do not cancel in the annual means. This effect is primarily attributed to the non-linear relationship between AoA and  $N_2O$ . This is explained by the following: 1) AoA strongly depends on the speed of the BDC, with lower AoA values indicating an acceleration, and higher AoA indicating deceleration of the vertical transport. In the absence of significant photolytic loss of  $N_2O$  via the Reaction (R7), the changes in stratospheric  $N_2O$  would be controlled only by changes of the rate of the tropical upwelling of the BDC (or simply by AoA), i.e. faster upwelling would enhance transport of  $N_2O$  to the stratosphere, and vice versa. Without photolytic loss, the rate of change of  $N_2O$  concentration would be inversely proportional to the AoA change; 2) the dominant chemical loss mechanism of  $N_2O$  is through its photolysis. The amount of photolysed  $N_2O$  depends on the residence time of  $N_2O$  and this in turn depends on the transport speed, i.e. AoA. Longer residence times of  $N_2O$  result from a transport slow-down. Consequently, there is more time for photolytical destruction of  $N_2O$ ; 3) as the amount of  $N_2O$  is controlled by both transport and photochemistry, its changes do not cancel in the annual average; 4) the amount of  $NO_2$  and  $O_3$  are chemically linked to that of  $N_2O$ . Overall, the changes of  $NO_2$  and  $O_3$  are dependent on both the amount of  $N_2O$  transported to the stratosphere and its residence time'.

Supplement – The notation of SCIAMACHY, TOMCAT, and Insignificant is confusing, since the gray Insignificant lines can be either of the former two. The current notation obscures the important fact that the subdivided SCIAMACHY measurements never show a significant trend in the opposite direction to the overall trend in  $N_2O$  and  $O_3$ . I recommend using just green, blue, and, if the authors think it is helpful, a dotted version of these colored lines for an insignificant trend.

We reworked Fig. S4-S7, i.e. we plotted statistically significant (2-sigma) changes as solid lines, and insignificant changes as dashed lines.

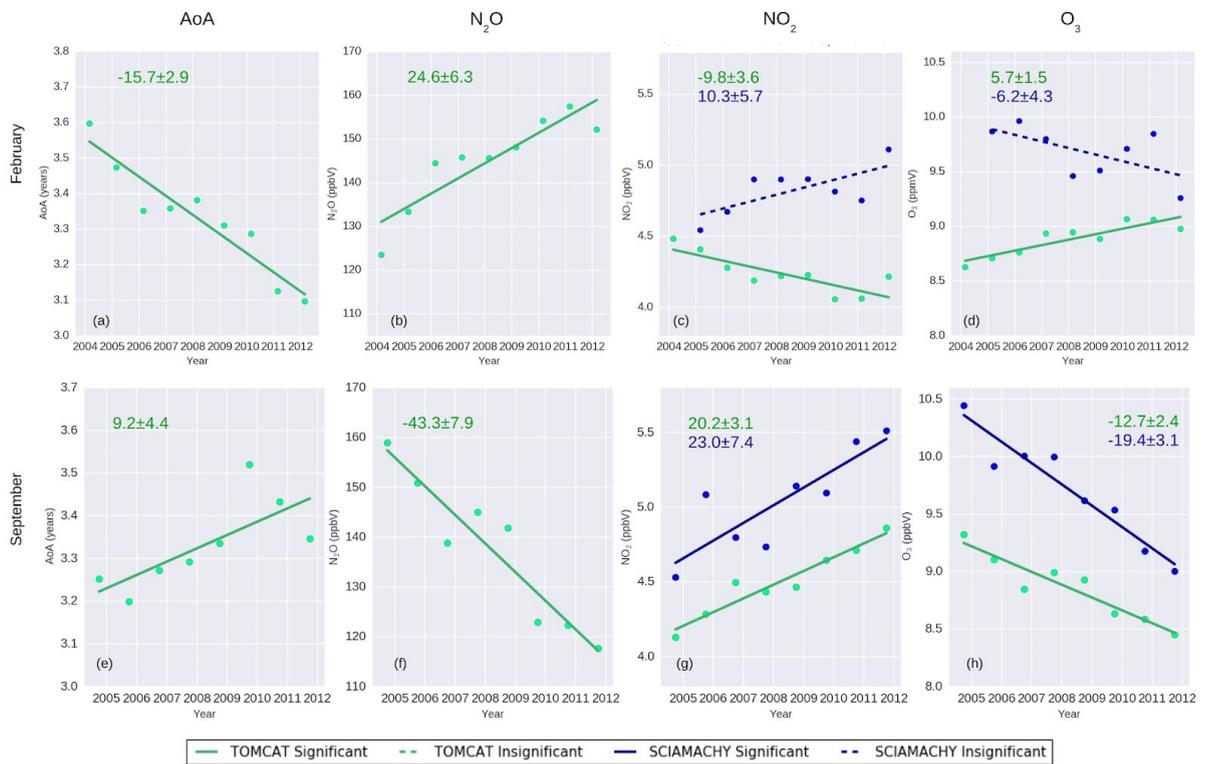


Figure 12. Linear changes of AoA,  $N_2O$ ,  $NO_2$ , and  $O_3$  minus QBO effect averaged over (a-d) Februaries 2004-2012 and (e-h) Septembers 2004-2011 in the tropical stratosphere between 30 and 35 km altitude. Colour coding indicates the data source: TOMCAT CNTL simulation (green), and SCIAMACHY measurements (dark blue). Colour-coded trend values and their errors (in % per decade) are shown in each panel. Solid lines indicate statistically significant linear changes at the  $2\sigma$  level, dashed lines indicate statistically insignificant changes.