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Dynamically controlled ozone decline in the tropical mid-stratosphere observed by SCIAMACHY

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This document contains additional figures to support those presented in the manuscript.

Figure S1 and **S2** show latitude-altitude plots of NO_x (NO + NO_2) and the bulk of NO_y species (NO_x + HNO_3 + $2xN_2O_5$) linear changes in % per decade for the period January 2004–April 2012 from TOMCAT control run (CNTL, **a**), run with constant tropospheric mixing ratios of source gases (fSG, **b**), and run with annually repeating meteorology (fDYN, **c**). Data are shown in the latitude range from 60° S to 60° N, altitude range from 15 to 40 km. TOMCAT CNTL and fSG runs show positive changes for both NO_x (**Fig. S1a,b**) and NO_y (**Fig. S2a,b**) of around 10 and 5 % per decade respectively in the tropical mid-stratosphere and are very similar in shape. TOMCAT fDYN run shows also positive changes for both NO_x (**Fig. S1c**) and NO_y (**Fig. S2c**) of around 3-5% per decade, but the shape of such positive change is different.

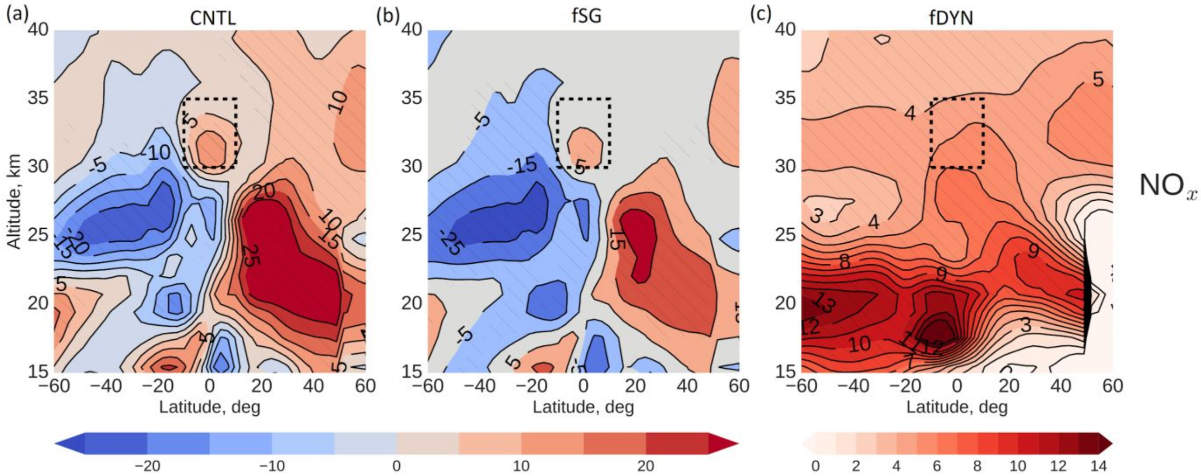

Figure S1. Latitude-altitude distribution of NO_x changes in % per decade from MLR model applied to TOMCAT runs for January 2004–April 2012 period: **(a)** CNTL, **(b)** fSG, and **(c)** fDYN. Hatched areas show significances at the 2σ level. Dashed rectangle indicates the region of tropical mid-stratosphere.

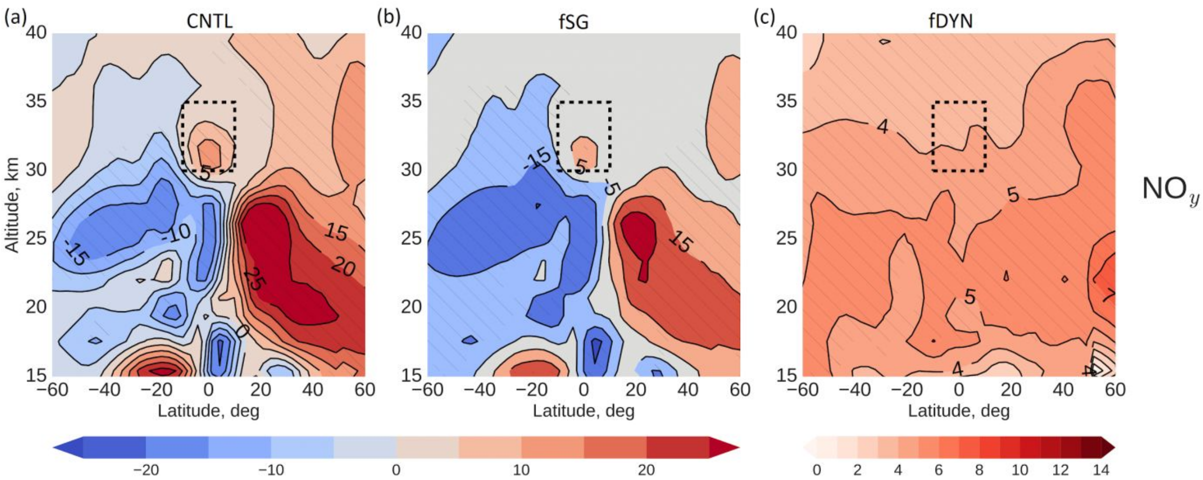

Figure S2. Latitude-altitude distribution of NO_y changes in % per decade from MLR model applied to TOMCAT simulations for January 2004–April 2012 period: **a)** CNTL, **(b)** fSG, and **(c)** fDYN. Hatched areas show significances at the 2σ level. Dashed rectangle indicates the area of tropical mid-stratosphere.

Figure S3 shows N_2O-AoA anti-correlation coefficients as the function of the month at altitudes 32, 33, and 35 km. To overcome hemispheric dependencies and confirm shown by Fig. 9 (see manuscript) seasonal variations of anti-correlation, we use narrower tropical band (4° S -4° N, **a**) and split it to the southern (4° S-1° S, **b**) and northern (1° N-4° N, **c**) regions. The anti-correlation was calculated at native TOMCAT grid. The dashed horizontal lines indicate an arbitrary threshold of moderate anti-correlation, which was selected to be the value of -0.6. The seasonal variations of anti-correlation are very similar to those, shown in Fig. 9 (see manuscript). In the narrow tropical region (**a**) the AoA-N_2O anti-correlation has the lowest values during December-March. During the other months of the year, it is moderate and reaches the highest values (absolute value of around 0.9) during end of northern hemisphere (NH) summer (August) and autumn (September, October) months at all analysed altitudes. Very similar seasonal behaviour is also observed in the southern hemisphere (**b**) with the lowest anti-correlation during December-February (and March at 35 km), which corresponds to southern hemisphere (SH) summer. The strongest anti-correlation is observed during May-October, which corresponds to SH winter. In contrast, in the tropical region of the NH (**c**) a decrease in AoA-N_2O anti-correlation is observed during summer months (June-July).

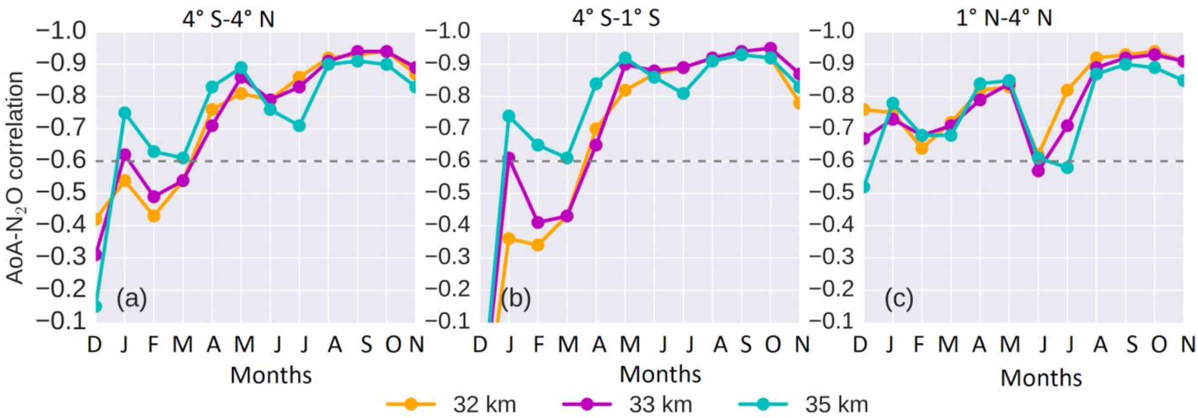

Figure S3. N_2O-AoA anti-correlation as the function of the month averaged for the period January 2004–April 2012 for (**a**) 4° S-4° N, (**b**) 4° S-1° S, and (**c**) 1° N-4° N. Colour coding indicates altitude: 32 km (orange), 33 km (magenta), 35 km (cyan). The dashed horizontal lines indicate the lower edge of moderate correlation, which was selected to be the value of -0.6.

Figures S4-S7 show linear changes of AoA, N_2O, NO_2, and O_3 minus QBO effect at 10 and 30 hPa averaged over each month during January 2004-April 2012 in the tropical stratosphere between altitudes 31.5 and 35 km. Solid lines stand for statistically significant changes, dashed lines stand for statistically insignificant changes. Colour coding denotes data source: SCIAMACHY (dark blue), TOMCAT CNTL run (green). Note, SCIAMACHY measurements (of NO_2 and O_3) always show higher concentrations in the tropical mid-stratosphere than TOMCAT simulations. Therefore SCIAMACHY changes are always plotted above TOMCAT data. In the top of each panel, we provide the information of trend value and its error (both in % per decade) for TOMCAT and where applicable for SCIAMACHY.

Figure S4 shows AoA, N_2O, NO_2, and O_3 linear changes during December, January, and February during the period 2004-2012 in the tropical mid-stratosphere. Solid lines indicate statistically significant changes and dashed lines indicate insignificant changes in TOMCAT (green) and SCIAMACHY (blue). **Figure S4** shows significant AoA decrease in January and February which (via impacting N_2O) leads to O_3 increase as modelled by TOMCAT. Though, SCIAMACHY measurements do not show any significant changes in NO_2 and O_3 during these months. In contrast, SCIAMACHY shows NO_2 slight increase and O_3 decrease, which is very similar to the regime of autumn months (further shown in **Fig. S7**).

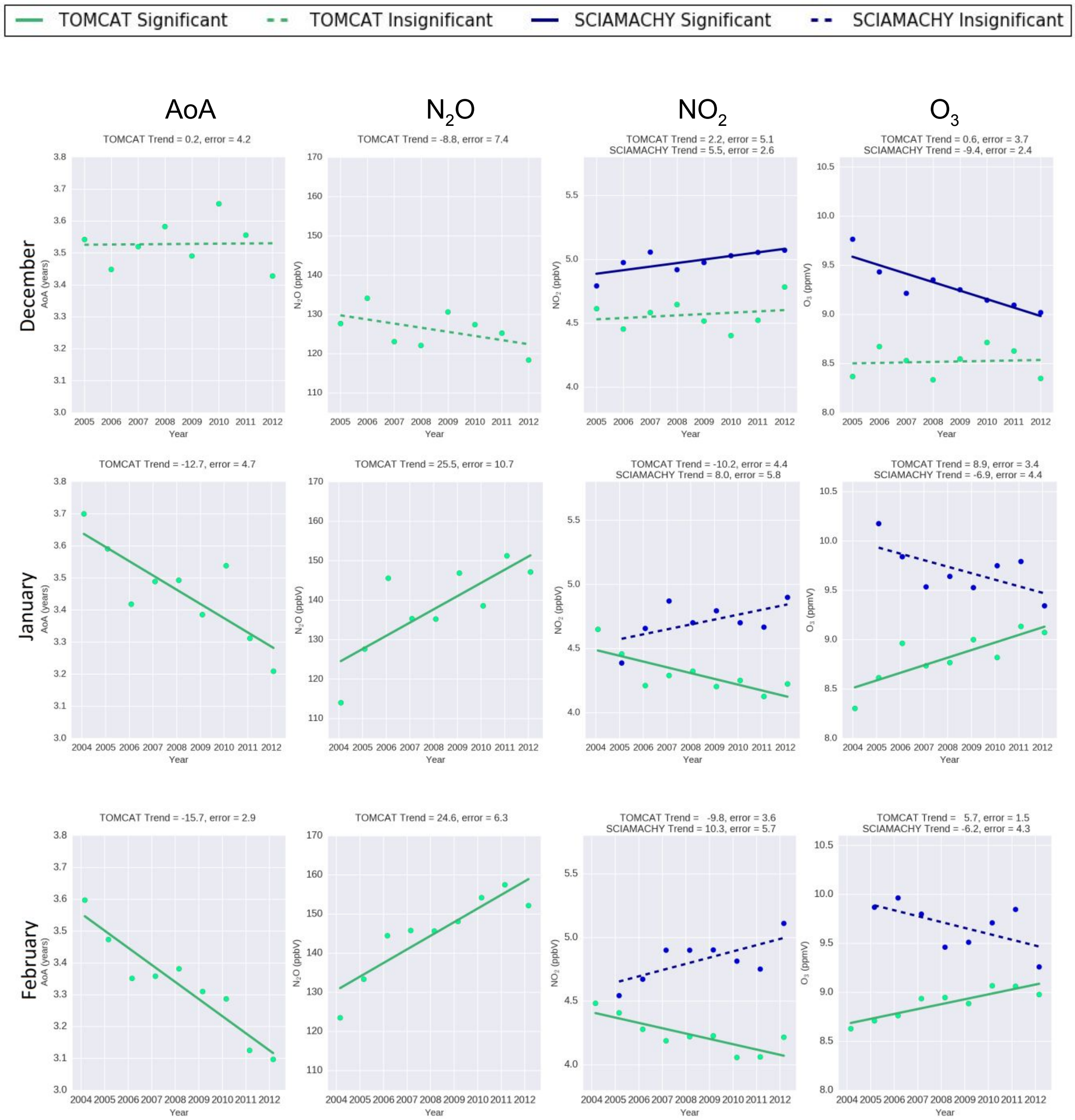

Figure S4. Linear changes of AoA, N_2O, NO_2, and O_3 minus QBO effect at 10 and 30 hPa in December, January, and February during the period 2004-2012 in the tropical mid-stratosphere. Solid lines stand for statistically significant changes, dashed lines stand for insignificant changes of TOMCAT (green) and SCIAMACHY (dark blue).

Figure S5 shows AoA, N_2O, NO_2, and O_3 linear changes during March, April, and May during the period 2004-2012 in the tropical mid-stratosphere. Solid lines indicate statistically significant changes and dashed lines indicate insignificant changes of TOMCAT (green) and SCIAMACHY (blue). **Figure S5** shows significant AoA decrease during all spring months as modelled by TOMCAT, but none of these changes finally lead to the increase of O_3.

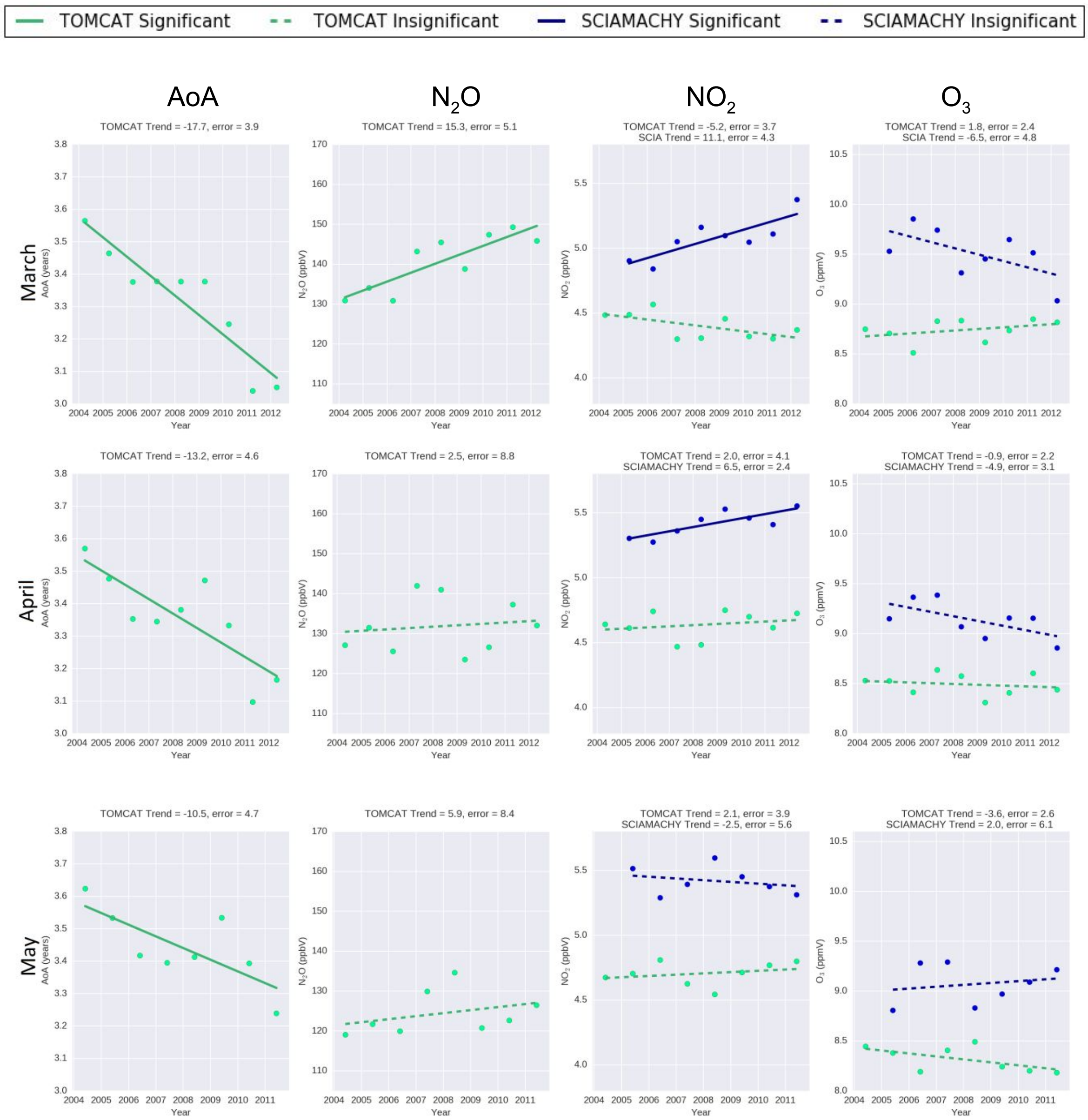

Figure S5. Linear changes of AoA, N_2O, NO_2, and O_3 minus QBO effect at 10 and 30 hPa in March, April, and May during the period 2004-2012 in the tropical mid-stratosphere. Solid lines stand for statistically significant changes, dashed lines stand for insignificant changes of TOMCAT (green) and SCIAMACHY (dark blue).

Figure S6 shows AoA, N_2O, NO_2, and O_3 linear changes in June, July, and August during the period 2004-2012 in the tropical mid-stratosphere. Solid lines indicate statistically significant changes and dashed lines indicate insignificant changes in TOMCAT (green) and SCIAMACHY (blue). **Figure S6** shows good agreement of O_3 decline between TOMCAT simulation and SCIAMACHY measurements in July and August. There are no statistically significant changes observed in June in N_2O, NO_2, and O_3 from model simulation and satellite measurements.

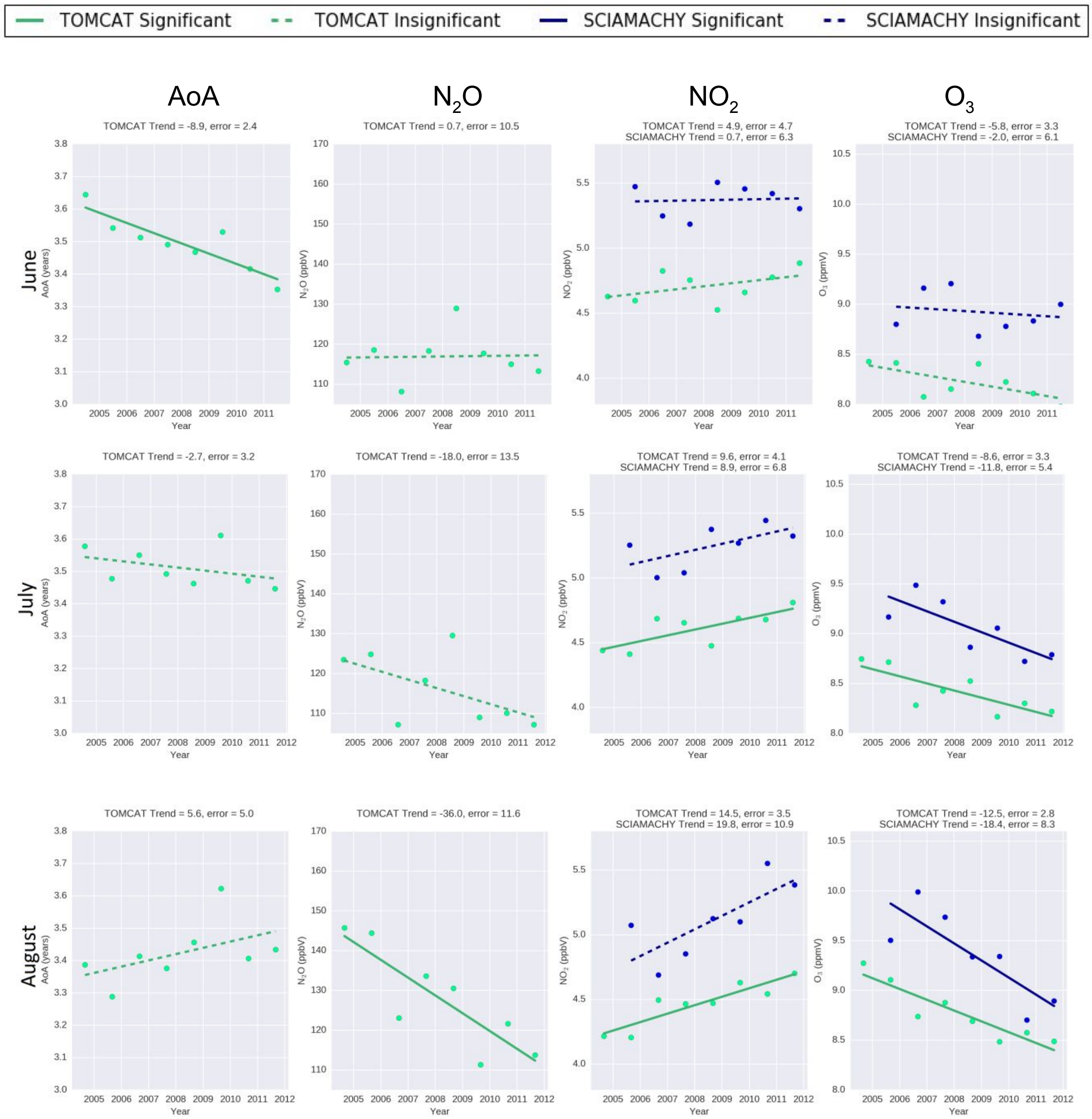

Figure S6. Linear changes of AoA, N_2O, NO_2, and O_3 minus QBO effect at 10 and 30 hPa in June, July, and August during the period 2004-2012 in the tropical mid-stratosphere. Solid lines stand for statistically significant changes, dashed lines stand for insignificant changes of TOMCAT (green) and SCIAMACHY (dark blue).

Figure S7 shows AoA, N_2O, NO_2, and O_3 linear changes in September, October, and November during the period 2004-2012 in the tropical mid-stratosphere. Solid lines indicate statistically significant changes and dashed lines indicate insignificant changes in TOMCAT (green) and SCIAMACHY (blue). **Figure S7** shows a significant increase of AoA during Septembers and Octobers which in the end caused strong O_3 decline. These changes agree very well between SCIAMACHY measurements and TOMCAT simulation. In November NO_2, and O_3 model and measurements also show good agreement, but no significant changes in transport were identified by the model.

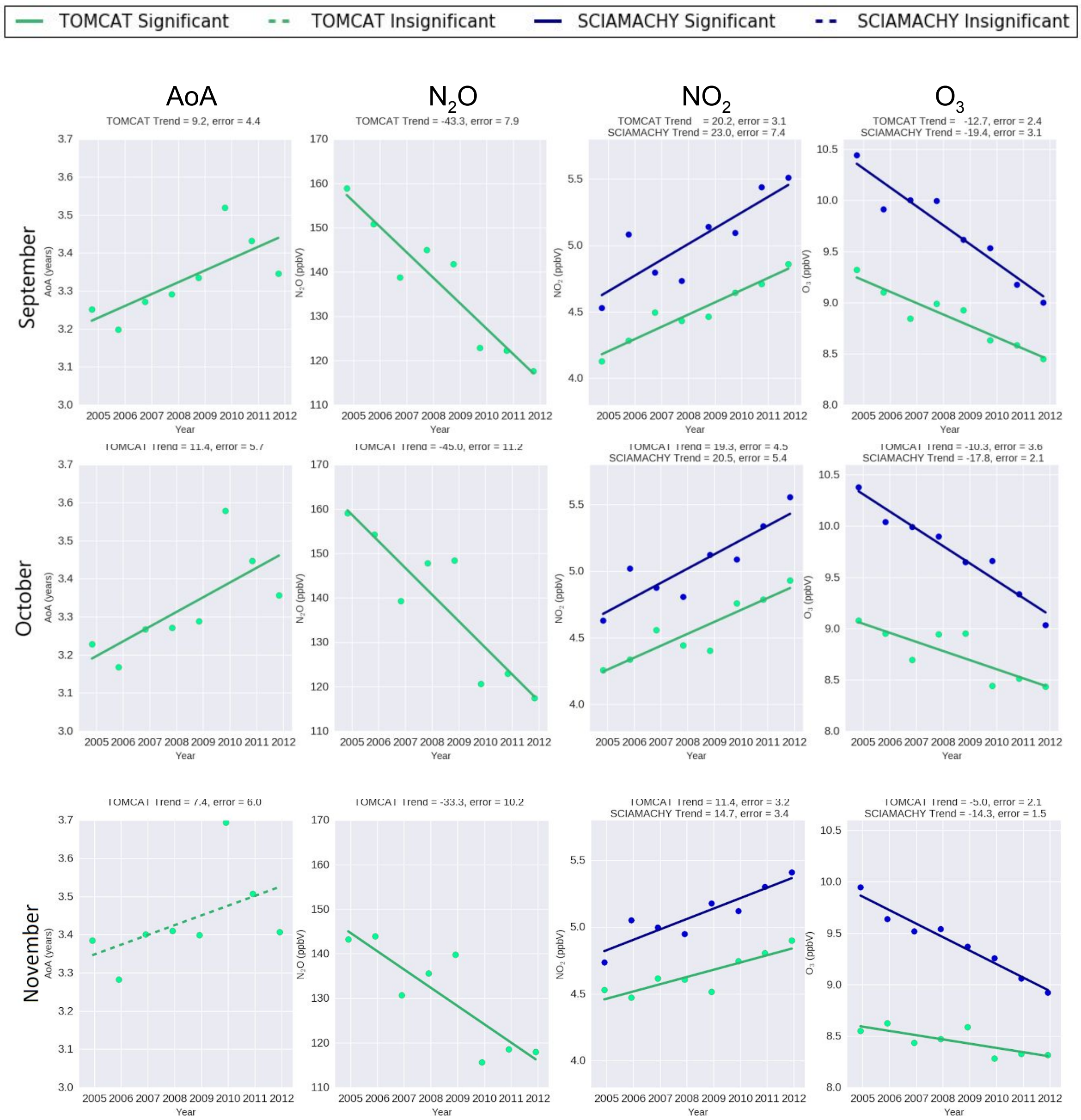

Figure S7. Linear changes of AoA, N_2O, NO_2, and O_3 minus QBO effect at 10 and 30 hPa in September, October, and November during the period 2004-2012 in the tropical mid-stratosphere. Solid lines stand for statistically significant changes, dashed lines stand for insignificant changes of TOMCAT (green) and SCIAMACHY (dark blue).