

## Response to Anonymous Referee #1

### Responses in red

#### 1 Overall comments

This paper highlights some results from 22 years of stratospheric ozone monitoring by the MOPI microwave radiometer at Lauder New Zealand, and puts them into the large scale context of global ozone and N<sub>2</sub>O measured by the MLS microwave radiometer on the AURA satellite. To me, the take home messages from the paper are

- Unusually high ozone in the mid-stratosphere (near 10 hPa) above Lauder in June 2001 was caused by anomalous transport associated with a strong Australian (stratospheric) anti-cyclone, that was unusually stationary above New Zealand throughout June 2001.
- Mid-stratospheric ozone at Lauder (near 10 hPa) shows high correlation with ozone throughout the Southern hemisphere mid-stratosphere (20\_S to 60\_S, 20 hPa to 3 hPa). It is also anti-correlated to tropical ozone (around 10 hPa, 10\_S to 10\_N), and to Northern hemispheric ozone (around 20 hPa, 20\_N to 50\_N). This correlation pattern appears to be caused by QBO modulation of trace gas transports.
- Enhanced ozone in the mid-stratosphere goes hand in hand with enhanced N<sub>2</sub>O. Enhanced N<sub>2</sub>O is usually associated with faster transport in the Brewer Dobson circulation, which leaves less time for photo-dissociation of N<sub>2</sub>O. Less photodissociation means less NO<sub>x</sub> and less chemical ozone destruction in the midstratosphere.
- Over the 2010 to 2013 period, ozone and N<sub>2</sub>O mixing ratios have been unusually low around 10 hPa in the tropics, and have been unusually high around 10 hPa in the Southern Hemisphere extra-tropics. The latitude altitude pattern of the 2004 to 2013 trend is very similar for ozone and N<sub>2</sub>O, and is very similar to the pattern associated with QBO-related ozone variations at Lauder.
- This anomaly during 2010 to 2013 suggests a slowing of ascent in the tropical mid-stratosphere, and faster transport to the Southern hemisphere mid- stratosphere. The most recent data since mid-2013 indicated that the transport anomaly has ended.

Overall I think this is a good paper and worth publishing in ACP. I do suggest a few minor revisions below.

#### 2 Comments on Text

The text does not always read well, and could benefit from some copy-editing.

page 5243, 1st paragraph. The importance of anthropogenic chlorine, increasing until the late 1990s, decreasing since 2000, should be mentioned here.

We have done quite a bit of copy editing throughout the manuscript. We also added some text about CFC's and the Montreal Protocol in the previous paragraph: "The large decline observed from the 1960s to the late 1990s has ended as a result of the reduction in chlorofluorocarbon (CFC) emissions following the 1987 Montreal Protocol (WMO, 2014)."

page 5243, 1st and 2nd paragraph. Rather than going paper by paper, it might be better to separate declining and increasing ozone trends, before and after 2000. What sticks out from this general picture is the unexpected ozone decline around 10 hpa since about 2002 in the tropics. This different decadal variability is the topic of the present paper!

We have rewritten this section of the paper in a more chronological, instead of instrument-based, order. This clarifies that all of the trends observed since the 1984-1997 Kyrola study of the SAGE measurements show the decline around 10 hPa in the tropics. The text now reads (the first paragraph below is unchanged), before going on to discuss SCIAMACHY:

“Several studies of satellite data show the variability in O<sub>3</sub> trends depending upon the exact timeframe and geographical location. Kyrölä et al. (2013), using measurements from the Stratospheric Aerosol and Gas Experiment (SAGE) from 1984-1997, show a general decrease in O<sub>3</sub> that is statistically significant over much of the stratosphere and is particularly large in the mid-latitude upper stratosphere. However, they also show an increase in equatorial O<sub>3</sub> (albeit not statistically significant) in the 30-35 km region.

There are a number of studies covering later years, all of which show a variation in O<sub>3</sub> that differs dramatically from that of the 1984-1997 SAGE data. Nedoluha et al. (2015) studied O<sub>3</sub> over the period 1991-2005, when Halogen Occultation Experiment (HALOE) measurements are available, and found a strong decrease in mid-stratospheric O<sub>3</sub> in the tropics over this period. Kyrölä et al. (2013) also examined the period 1997-2011, showing a general increase in O<sub>3</sub> from SAGE and Global Ozone Monitoring by Occultation of Stars (GOMOS) measurements, but a statistically significant decrease near 30 km in the tropics.”

Pages 5251 to 5253. This discussion is very detailed and hard to follow. Maybe shorten? Or provide more structure / subsections. E.g. start a new subsection "Links between Ozone and N<sub>2</sub>O" in line 11 of page 5251.

We have reorganized and rewritten parts of Section 4, and now divide it into 3 subsections

4.1 Monthly O<sub>3</sub> anomaly correlations

4.2 Links between O<sub>3</sub> and N<sub>2</sub>O

4.3 Decadal changes in O<sub>3</sub> and N<sub>2</sub>O

Section 4.1 has been rewritten to read:

“To better understand the global implications of the observed O<sub>3</sub> variations over Lauder, we investigated how the variations in O<sub>3</sub> observed over Lauder compare globally with changes in MLS O<sub>3</sub>. We first calculated monthly averaged O<sub>3</sub> at each MLS pressure level from 50 to 1 hPa in 2° latitude bins for 10 years of MLS data (2004-2014). We then calculated a climatological (i.e., 10-year) average for each calendar month. Using this climatology, we calculated an anomaly for each month of the 10-year series as a function of latitude and pressure. A similar monthly anomaly time series was calculated for the MOPI ozone at 10 hPa. Correlation coefficients were calculated between the 10 hPa MOPI anomalies and the MLS anomalies at different pressures and latitudes, using months where both MLS and MOPI measurements were available.

Figure 8 shows the correlation coefficient ( $r$ ) as a function of pressure and latitude. The strongest correlation occurs slightly equatorward of Lauder and at a slightly higher pressure level. This is likely due to differences in instrumental errors, vertical resolution, and because the MOPI measurement is for local conditions near Lauder and not a zonal average. At the equator and 10 hPa, there is a strong anti-correlation ( $r < -0.5$ ) between MOPI and MLS (the correlation between 10 hPa MLS O<sub>3</sub> at 45°S and the equator is similar). There is also a weaker anti-correlation between MOPI and MLS O<sub>3</sub> at ~20-45°N below 10 hPa.

The geographical correlations seen in Fig. 8 are similar to those discovered by Randel and Wu (1996), who used singular value decomposition (SVD) analysis to study the relationship between QBO zonal winds and global SAGE O<sub>3</sub> anomalies. The second mode of their analysis (SVD2; which explains 25% of the overall covariance) shows an anti-correlation between 10 hPa O<sub>3</sub> at Southern mid-latitudes and at the equator. It also shows a much weaker anti-correlation between Southern mid-latitudes and Northern mid-latitudes.”

We also did change a reference from Figure 5 to Figure 7 (old Figure 6) in 5251 line 1.

Start a new subsection "Decadal changes" on line 16 of page 5253?

Good idea. Done.

Page 5254. As mentioned briefly in the introduction, several papers remark on a 2002 to 2013 ozone decline in the tropics near 10 hPa. There is also a recent modelling study by Aschmann et al. (2014) trying to explain decadal variations in tropical ozone. I think some additional discussion is needed in the conclusions section (or before) to put the results from the current paper into this wider context.

The Aschmann study is quite interesting, but it is for slightly lower altitudes (70-30 hPa), where things seem to develop differently than at 10 hPa in the tropics, or at lower altitudes in mid-latitudes. We have added a reference and some discussion related to Mahieu et al. [2014] since, at least for the mid-latitudes, our Figure 9 shows a change in sign in N<sub>2</sub>O consistent with their conclusions relating to age-of-air.

### 3 Comments on Figures

I think it would make sense, to plot satellite data also in at least one of the panels of Figure 3.

We plot the individual monthly MOPII points to give an idea of the typical variation in the monthly average. If we plot satellite data on here as well it would become difficult to convey that information.

Are all panels of Figure 3 necessary? The 10 hPa panel would probably be enough.

We provide 4 panels in order to show the levels at which our two features of interest are most clearly observed, and to visually make the point that while the 2009-2013 anomaly occurs in the mid- to upper stratosphere, the 2001 anomaly is primarily a lower stratospheric phenomenon.

Is Figure 5 necessary? The same information could be incorporated into Figure 3, and more or less appears again in Figure 6. I think Figure 5 could be dropped.

Figure 5 (now Figure 6) differs from Figure 6 (now Figure 7) in 3 primary ways: 1) It shows the seasonal variation (there seems to have been some confusion on what the following figure shows, which we have hopefully clarified). 2) It shows that the difference between convolved and unconvolved Aura MLS data (which is small), and 3) it shows the Aura MLS measurements with a much tighter coincidence to Lauder. Despite all of these differences Figure 5 still shows clearly the increase in ozone in 2009/2010, so it helps to emphasize that this increase can be clearly detected without taking annual averages.

Figure 6: This is quite a good Figure. I don't understand, however, if seasonal means

or annual means are plotted. The caption says annual means, but the lines indicate seasonal means. Please correct / clarify.

These are annual means shown 4 times yearly. We have added within the parentheses of the caption the phrase “with annual averages taken from January-December ...” to hopefully clarify this point.

Figure 9: I think it would be good to also give an indication of the significance of these decadal trends. Looking at Figure 8, it seems to me that the trend is only a small part of the overall variance, and uncertainties in the QBO part of the fit will also result in large uncertainties in the linear trend. Although insignificant trends can indicate a consistent change, they have less meaning for the long-term evolution.

We now provide, in the text, average and maximum uncertainties for the trends at the pressures shown in the figure (both at Lauder and globally). The text now reads: “Based on the monthly MLS dataset that was used for the fit, the average 1- $\sigma$  uncertainty in the O<sub>3</sub> (N<sub>2</sub>O) trend fit is 0.008 ppmv/yr (0.46 ppbv/yr), and it is <0.020 ppmv/yr (<1.05 ppbv/yr) everywhere in Fig. 10. The 1- $\sigma$  uncertainty in the O<sub>3</sub> (N<sub>2</sub>O) trend fit at 45°S is <0.011 ppmv/yr (<0.76 ppbv/yr).”

#### **4 Reference**

Aschmann, J., Burrows, J. P., Gebhardt, C., Rozanov, A., Hommel, R., Weber, M., and Thompson, A. M.: On the hiatus in the acceleration of tropical upwelling since the beginning of the 21st century, *Atmos. Chem. Phys.*, 14, 12803-12814, doi:10.5194/acp-14-12803-2014, 2014.