

5



# On ozone trend detection: using coupled chemistry-climate simulations to investigate early signs of total column ozone recovery.

James Keeble<sup>1</sup>, Hannah Brown<sup>1</sup>, N. Luke Abraham<sup>1,2</sup>, Neil R. P. Harris<sup>3</sup>, and John A. Pyle<sup>1,2</sup>

<sup>1</sup>University of Cambridge, Department of Chemistry, Cambridge, UK

<sup>2</sup>National Centre for Atmospheric Science, Cambridge, UK

<sup>3</sup>Centre for Environmental and Agricultural Informatics, Cranfield University, Cranfield, UK

Correspondence to: James Keeble (james.keeble@atm.ch.cam.ac.uk)

Abstract. Total column ozone values from an ensemble of UM-UKCA model runs are examined to investigate different definitions of progress on the road to ozone recovery. This approach takes into account the internal atmospheric variability

- 10 of the model in assessing the statistical significance of each definition. Three definitions of recovery are investigated: (i) a slowed rate of decline and the date of minimum column ozone; (ii) the identification of significant positive trends; and (iii) a return to historic values. A return to past thresholds is the last state to be achieved. However, while recovery may appear to be robust at a particular point of time, additional years of observations may lead to a reduced significance of trends due to natural variability (e.g. solar cycle, QBO, ENSO). This points to the need to ensure that the impact of natural cycles on total
- 15 ozone is correctly described in statistical models, especially in the tropics where chemical depletion of the column is small. Trends for the 2000-2017 period are positive at most latitudes and are statistically significant when natural cycles are accounted for. This significance results largely from the large sample size of the multi-member ensemble. The influence of the natural cycles on trend determination is least at latitudes where the trends are sizeable. Thus, while ozone recovery can be identified in certain months over Antarctica, the mid-latitudes are the best place to identify early recovery as the trends
- 20 are large compared to the variability. Over the Arctic, total column ozone is too variable for a signal to be easily detected: this arises both from the large dynamical interannual variability and from the large changes in chemical ozone loss from year to year. In the tropics, trends are too small compared to the natural variability to identify any statistical significance.

## 1 Introduction

2017 marks the 30<sup>th</sup> anniversary of the Montreal Protocol, which was implemented to protect the stratospheric ozone layer 25 from the harmful effects of ozone depleting substances (ODS). These gases, mostly inert in the troposphere, were shown to breakdown when they reached the stratosphere, with the subsequent products then leading to chemical ozone depletion (e.g. Molina and Rowland, 1974; Stolarski and Cicerone, 1974; Rowland and Molina, 1975). Controls introduced under the Montreal Protocol and its subsequent amendments slowed the rate of accumulation of these halogenated ODSs in the atmosphere, and since the late 1990s their atmospheric concentrations have begun to decline (Newman et al., 2006; Mäder et

- 30 al., 2010; WMO 2011; 2014). A reduction in equivalent stratospheric chlorine (ESC; Eyring et al., 2007) concentrations should lead to an increase in atmospheric ozone as the strength of the halogen catalyzed ozone destruction cycles declines. However, detecting recovery of the stratospheric ozone layer is complicated by a number of additional factors which affect the year to year variability of total column ozone values. These factors include volcanic eruptions, such as the eruption of Mt. Pinatubo in 1991 (e.g. Randel and Wu, 1995; Telford et al., 2009), changes in the solar cycle (e.g. Brasseur, 1993; Van
- 35 Loon and Labitzke, 2000; Austin et al., 2007; Calisesi and Matthes, 2007) and variability in ozone resulting from a range of factors affecting dynamical variability, including the quasi-biennial oscillation (QBO; e.g. Hollandsworth et al., 1995; Baldwin et al., 2001; Leblanc and McDermid, 2001) and the El Niño-Southern Oscillation (ENSO; e.g. Randel et al., 2009; Manzini, 2009). In addition, long term total column ozone trends are driven in part by emissions of other, non-chlorinated





anthropogenic species, such as  $CO_2$ ,  $CH_4$  and  $N_2O$ , which affect stratospheric ozone concentrations by altering stratospheric temperatures and dynamics (Haigh and Pyle, 1982; Plumb, 1996; Avallone and Prather, 1996; Eyring et al., 2010; 2013; Iglesias-Suarez et al., 2016), and in the case of  $CH_4$  and  $N_2O$  by acting as source gases for reactive  $HO_x$  and  $NO_x$  species (Chipperfield and Feng, 2003; Ravishankara et al., 2009; Revell et al., 2012; Meul et al., 2014). Identification of significant

- 5 trends is also made problematic by the difference in year-to-year variability in total column ozone values in different regions. For example, high northern latitudes exhibit very large interannual variability in winter and spring, while variability in the southern hemisphere is comparatively smaller. Furthermore, there is a dynamical response to changes in chemical ozone depletion in the stratosphere, which may enhance/impede future recovery by altering the transport of ozone (e.g. McLandress et al., 2011; Braesicke et al., 2013; Keeble et al., 2014; Solomon et al., 2016). As a result of all of these factors, identifying
- 10 robust recovery of total column ozone and ascribing that recovery to a decline in stratospheric halogen species is a complex issue.

Recovery of the stratospheric ozone layer could be detected using two different methodologies. For past trends, observations can be compared with a detailed chemistry-transport model which includes all known processes. Good agreement between the model and observations when all processes are included, but not when polar halogen chemistry is included and

- 15 interannual dynamical variability is excluded, could constitute a sign of ozone recovery (e.g. Solomon et al., 2016). Alternatively, a statistical approach can be followed in which data are used to detect significant change between time periods. The impact of confounding changes (e.g., QBO, solar cycle, etc.) can be quantified using multiple linear regression and removed from the statistical analysis of the data in order to provide a better estimate of long-term trends (e.g. Staehelin et al., 2001; Reinsel et al., 2005; WMO, 2007; Harris et al., 2015; Chipperfield et al., 2017). These statistical approaches
- 20 nearly all work by relying on the somewhat simplistic assumption of a linear relationship between a proxy variable and its impact on total ozone.

To explore future ozone trends and recovery, data from fully coupled chemistry-climate model (CCM) simulations are required. Each CCM simulation constitutes a possible future evolution of stratospheric ozone. In order to sample the effect of internal atmospheric variability on ozone and to derive an estimate of uncertainty of future trends, multiple ensemble

- 25 members can be run in which the initial conditions of each simulation are modified but the same forcings are prescribed (e.g. GHG (greenhouse gas) evolution, SSTs, aerosol loadings). Greater confidence can be assigned to significance of the mean trend as the number of ensemble members increases. Multiple ensemble members also give information about the possible range of future trends. Thus, using an ensemble of future projections from a single CCM can provide additional insight into the detection of different phases of ozone recovery.
- 30 In this study, we use results from a chemistry-climate model coupled with statistical approaches to explore different definitions of ozone recovery (see Reinsel et al., 2005; Weatherhead and Andersen, 2006; Chipperfield et al., 2017). In particular we define three stages of total column ozone recovery:
  - 1. A reduced rate of decline in ozone and the date of minimum ozone.
  - 2. Statistically significant increases in column ozone values that can be ascribed to reductions in ESC.
- 35 3. Return of total column ozone values to some specified past value (typically 1980 or 1960).

Identifying when and if each of these stages has occurred at different latitudes – and being able to assess the confidence with which this can be done - is fundamental to determining the success of the Montreal Protocol. For this work we use the ozone fields calculated in an ensemble of UM-UKCA transient simulations, described briefly in section 2, as proxies for atmospheric observations. We carry out a statistical analysis of the model results, as outlined in section 3, to identify when





each of these stages of recovery occurs for different latitude ranges. These results are presented in section 4, 5 and 6 and implications are discussed in section 7.

#### 2 Model configuration and simulations

An ensemble of transient simulations was performed using version 7.3 of the HadGEM3-A configuration of the Met Office's
Unified Model (Hewitt et al., 2011) coupled with the United Kingdom Chemistry and Aerosol scheme (hereafter referred to as UM-UKCA). This configuration of the model has a horizontal resolution of 2.5° latitude by 3.75° longitude, and 60 vertical levels following a hybrid sigma-geometric height coordinate with a model top at 84 km. The chemical scheme used in this configuration of the model is an expansion of the scheme presented in Morgenstern et al. (2009) in which halogen source gases are considered explicitly, resulting in an additional 9 species, 17 bimolecular and 9 photolytic reactions.

- 10 Stratospheric aerosol concentrations are prescribed using a climatology based on observations (from SPARC, 2006; described by Eyring et al., 2008) for the historical part of the run, after which background concentrations of stratospheric aerosol loadings are prescribed. HadGEM3-A includes an internally generated quasi-biennial oscillation (QBO), which in this configuration of the model has a period of ~27 months while the magnitude of modelled easterly(westerly) equatorial zonal wind speed is ~25 m s<sup>-1</sup> (10m s<sup>-1</sup>), both aspects in good agreement with observed zonal winds at Singapore (e.g. Lee
- 15 and Smith, 2003). The configuration of the model used for this study includes the effects of the 11-year solar cycle in both the radiation and photolysis schemes. The top of atmosphere solar flux follows historical observations from 1960 to 2012, after which a climatological solar cycle is imposed which is an average of the four solar cycles preceding 2012.

The transient simulations were performed following the experimental design of the WCRP/SPARC CCMI REF-C2 experiment (Eyring et al., 2013), which adopts the RCP6.0 scenario for future GHG and ODS emissions. Two of these

- 20 ensemble members were run from 1960 to 2099 and an additional five were run from 1980 to 2080. All ensemble members have identical time-dependent boundary conditions, but differ in their atmospheric initial conditions, thereby providing an estimate of internal atmospheric variability. The simulations were performed in an atmosphere-only configuration, and each ensemble member uses prescribed sea surface temperatures (SSTs) and sea ice fields taken from a parent coupled atmosphere-ocean HadGEM2-ES integration as lower boundary conditions. The simulations used for this study are
- 25 described in more detail in Bednarz et al. (2016) and Keeble et al. (2017), and were performed in support of phase 1 of the Chemistry–Climate Model Initiative (CCMI; Morgenstern et al., 2017).

#### **3 Removing natural cycles**

Identifying an increase in total column ozone resulting from reductions in stratospheric chlorine requires removing the effects of natural processes, such as volcanic eruptions, the QBO, ENSO and solar cycle, from the modelled total column ozone data, as these cycles may impose short terms trends in the data which are wrongly interpreted as signs of recovery. In order to identify the impacts of these natural processes on modelled total column ozone we create a statistical model using multiple linear regression (MLR) analysis. This process assumes that the modelled total column ozone values can be reproduced by combining some constant value of total column ozone TO3<sub>*i*</sub>, which corresponds to the intercept term of the MLR, with a number of explanatory, or predictor, variables. This statistical model can be expressed as:

$$TO3_{e,l,t} = TO3_{e,l,i} + \alpha^{QBO_{50}} QBO_{50_{e,t}} + \alpha^{QBO_{30}} QBO_{30_{e,t}} + \alpha^{solar} . solar_t + \alpha^{ENSO} . ENSO_t + \alpha^{aerosol} . aerosol_t + \alpha^{ESC} . ESC_{e,l,t} + N_{e,l,t}$$





in which the  $\alpha$  values are the coefficients returned from the MLR for each explanatory variable (denoted by the superscript) and vary between latitude range and ensemble member. The explanatory variables included in the MLR are the QBO, solar cycle, ENSO, volcanic aerosols and ESC. For the QBO, two terms are included,  $QBO_{50}$  and  $QBO_{30}$ , which correspond to equatorial westerly winds at 50 hPa and 30 hPa respectively. Two QBO terms are included to account for the phase shift in

- 5 the total column ozone response with respect to QBO changes at different altitudes. The solar cycle is represented by the top of atmosphere solar flux, represented in the MLR as *solar<sub>t</sub>*. ENSO effects on column ozone are represented by *ENSO<sub>t</sub>*, the detrended sea surface temperature anomalies in the NINO3.4 region. Volcanic aerosols are included as hemispheric aerosol optical depths, and so are different for the northern and southern hemispheres to account for the lack of interhemispheric transport of aerosols emitted into the stratosphere from high latitude eruptions. The final term included in the MLR, *ESC*,
- 10 represents stratospheric chlorine concentrations. This term is equal to the ESC concentration at 30 km for each latitude bin to account for the time taken for ODS to be transported to higher latitudes. Any noise which is not explained by the MLR is represented by the term *N*. The subscripts *e*, *l* and *t* indicate that the explanatory variable differs with ensemble member, latitude and/or time respectively. *Solar*<sub>t</sub>, *ENSO*<sub>t</sub> and *aerosol*<sub>t</sub> terms are all prescribed forcings in the model and do not vary between ensemble members.
- 15 This statistical model can then be used to remove the component of total column ozone variations related to the QBO, solar cycle and volcanic aerosol changes from the raw model data to leave a set of ozone residuals which retain the long-term trend and any interannual variability not explained by the MLR:

$$RO3_{e,l,t} = TO3_{e,l,t} - \left(\alpha^{QBO_{50}}, QBO_{50e,t} + \alpha^{QBO_{30}}, QBO_{30e,t} + \alpha^{solar}, solar_t + \alpha^{ENSO}, ENSO_t + \alpha^{aerosol}, aerosol_t\right)$$

#### 4 Modelled global column ozone and minimum values

- Figure 1 shows deseasonalised monthly mean total column ozone anomalies relative to 1980 values, averaged over 60°S-20 60°N, from 1960 to 2100 for each individual ensemble member (light blue lines) and the ensemble mean (dark blue line). A sharp decrease in total column ozone is modelled from 1980 to the late 1990s, consistent with increased ESC loadings resulting from the use and emission of ODS. From the late 1990s until ~2070 column ozone values gradually increase, exceeding their 1980s values by ~2030, and their 1960s values by ~2050. Beyond 2070 total column ozone values remain relatively constant until the end of the century. Superimposed on these long term trends is the effect of the solar cycle,
- 25 which imprints a distinctive 11-year oscillation in the data. Alongside the modelled total column ozone anomalies are shown values from version 2.8 of the Bodeker Scientific total-column ozone dataset in black (Bodeker et al., 2005). There is generally good agreement between modelled total column ozone anomaly values and the Bodeker dataset; decadal total column ozone changes, the response of column ozone to the solar cycle and the magnitude of interannual variability are all well captured by the model throughout the time period during which the observations and model data overlap.
- 30 Also shown in Figure 1 are the ozone residuals calculated when the effects of natural cycles are removed, as detailed in section 3 (red lines). This dataset follows the long term trends of the raw UM-UKCA data, but the cyclic short term trends in column ozone values have been removed. Most obviously from Figure 1 is the removal of the 11-year solar cycle signal, leading to a much smoother, monotonically increasing trend from 2000 to 2060 compared to the raw model data.

As discussed above, the first signs of detectable ozone recovery would be identified as a reduced rate of decline in column 35 ozone and the date of minimum ozone. Modelled total column ozone values generally decrease from 1980 to the late 1990s (blue line, Figure 1), consistent with the increase in ESC amounts. However, this decrease is not constant; rapid decline is modelled from 1980 to 1985 and from 1990 to 1995, while between these periods total column ozone abundances are





relatively constant or even increase (see inset in Figure 1). This feature is also seen in the Bodeker dataset, and predominantly results from the impact of the solar cycle on stratospheric ozone concentrations. As top of atmosphere solar flux decreases from solar maximum to solar minimum, rapid decline of total column ozone occurs as this effect combines with the impacts of increasing ESC. Conversely, as top of atmosphere solar flux increases, enhanced stratospheric ozone

- 5 production temporarily offsets the chemical ozone destruction resulting from increased ESC concentrations. This is confirmed by analyzing the ozone residuals (red line Figure 1), which show a much smoother decline from 1980 to the late 1990s, and highlights the importance of understanding the drivers of short term trends in raw total column ozone values when trying to assess longer term trends.
- As well as influencing the trajectory of declining column ozone abundances, natural cycles also affect the timing and 10 magnitude of minimum total column ozone values. In the raw model data, the minimum total column ozone values averaged over 60°S-60°N are reached between 1992-1994, depending on the ensemble member, which is several years before the peak loading of ESC in 1997 (e.g. Mäder et al., 2010; WMO 2011; 2014). This offset in timing between peak ESC and total column ozone minima results from the impact of the solar cycle, as discussed above, and the eruption of Mt. Pinatubo on total column ozone. The early 1990s was a time of low top of atmosphere solar flux, while the eruption of Mt. Pinatubo
- 15 increased stratospheric sulphate surface area density, both reducing total column ozone abundances. When the effects of these natural cycles are removed (red line Figure 1) total column ozone values are seen to be greater throughout the early 1990s.

Although this work indicates minimum column ozone values occurred in the 1990s, this is a poor metric for making robust conclusions about ozone recovery. Firstly, the ozone minimum may occur because there is no more capacity for increased chemical depletion despite increasing ESC. This is the case over Antarctica during springtime during the 1990s, where near complete destruction of polar lower stratospheric ozone occurs and any additional increase in ESC would have a negligible effect. Secondly, minimum column ozone values are very sensitive to dynamical conditions. For example, Bednarz et al.

(2016) have shown that even under much lower stratospheric halogen loadings significant ozone depletion can occur in the

Arctic lower stratosphere during conditions which favour a cold, stable polar vortex.

# 25 5 Regional trends

Decline and subsequent recovery of total column ozone is often calculated using piecewise linear trends in two periods either side of an inflection time (e.g. Newchurch et al., 2003; Reinsel et al., 2005; Jones et al., 2009; Nair et al., 2013; Chehade et al., 2014). Previous studies have identified 1997 as the inflection time for long-term total column ozone observed trends (e.g. Harris et al., 2008), and as a result we define the decline phase as 1980-1997 with the recovery phase defined from

30 2000-2017. Here we calculate trends for both the decline and recovery phases firstly using the raw total column ozone data from the UM-UKCA model (discussed below) and then using model data in which the effects of the natural processes discussed above have been removed using the statistical model introduced in Section 3.

Figure 2 shows DU year<sup>-1</sup> trends for the decline (1980-1997) and recovery (2000-2017) phases for the column ozone data from the UM-UKCA simulation and the ozone residuals when natural cycles are accounted for, averaged over 10° latitude

35 bands. Error bars associated with each trend represent the 95% confidence intervals. During the decline phase, ozone trends from both datasets are greatest at high latitudes due to the heterogeneous activation of chlorine on PSCs within the polar vortex. The uncertainty associated with these trends is also largest at high latitudes, due to the higher year-to-year variability in chemical and dynamical processes at high latitudes compared with the tropics. Trends in the raw column ozone data are significant at all latitudes, although when natural cycles are removed the trends from 10°S-10°N are not significant. At all





latitudes there is a more negative trend in the raw UM-UKCA data compared with the dataset in which the natural cycles have been removed. This is the result of the pronounced solar minima during the 1990s, which resulted in lower column ozone values and so a greater trend from 1980. This can be clearly seen in Figure 1 by comparing the blue and red lines.

When considering the recovery phase, positive trends are modelled at all latitudes from 2000-2017. These trends are only

5 significant at the 95% confidence interval in the southern hemisphere between 80°S-50°S. However, when the effects of natural cycles are removed from the data, significant trends can be identified in the southern hemisphere between 80°S-30°S, and for the first time in the northern hemisphere from 20°N -70°N. For both datasets significant trends cannot be identified by 2017 at the highest latitudes due to the large year to year variability in the data

Identification of significant trends depends on the gradient of the trend, the number of data points (in this case the number of modelled monthly means) and the variance of the data. Analyzing the near global (60°S-60°N) raw total column ozone data (blue lines, figure 1), year 2000 is a solar maximum year and so total column ozone values are relatively high compared to the following few years. It is not until ~11 years later, during the next solar maximum, that trends become positive. Trend analysis on data between 2000 and 2015 could indicate that there is a significant positive trend, which could in turn lead to the conclusion that significant recovery of the ozone layer had begun. However, as further years are considered, from 2015

15 to 2020, total column ozone values start to decline as the solar cycle moves towards a solar minima, and the magnitude of the recovery trend is reduced while the variance in the residuals increases. Now, trends calculated from 2000 to 2020 are no longer statistically significant. As a result, when assessing recovery trends it is necessary to use datasets in which the effects of natural cycles have been accounted for, such as the ozone residuals calculated in section 3.

Figure 3 shows the month after 2000 at which trend significance can be identified in the ozone residuals for either the first 20 time (blue), or final time (red). Here trends are identified as significantly different from zero when they have a p-value < 0.05. Starting from 2000, the first month trends become significant (initial recovery) and the month after which they remain significant (robust recovery) can be calculated for each ensemble member. Distinguishing between these initial significance and robust significance dates is necessary since, as discussed above, trends can be significant after a number of months and then become non-significant as more data is added which increases the variance in the data or decreases the magnitude of the

- 25 trend. The first instance of detection of significant trends can be considered as false recovery if it does not coincide with the time after which trends never become non-significant. This is most clearly exhibited in the raw model data when considering the solar cycle, as discussed above, but is also true for the ozone residuals. However, when natural cycles are removed, trend estimates become significant earlier due to the reduced variance in the data, and both initial and robust recovery occur closer together. Note that, if the MLR described in section 3 accurately represented all drivers of interannual
- 30 variability (i.e. the *N* term was zero), there would be no distinction between initial and robust recovery.

Mid latitude trends become significant earlier than those of the tropics or high latitudes. This is due to the high degree of interannual variability at high latitudes, particularly in the Arctic, and the small magnitude of the trends in the tropics. Therefore, it is likely that both initial and robust recovery will first be observed in the mid latitudes. In addition, both measures of recovery occur at similar times, minimizing the risk of identifying false recovery. Correct identification of

35 robust recovery is important when considering observations of total column ozone and highlights the need to treat detection of significant recovery for the first time with caution as additional months/years of observational data may reduce the statistical significance of any observed trends.





#### 6 Return to historic values

While identification of statistically significant increases in total column ozone is a real sign that ozone recovery is occurring, recovery can be said to be complete when column ozone values reach their pre-CFC values again. Traditionally these return thresholds are taken to be either 1980 or 1960 values; here we use 1980. It is likely that total column ozone values will

5 initially exceed the 1980s threshold and then fall below this value again due to interannual variability and the effects of the solar cycle and QBO. As a result two metrics are considered: the first time total column ozone exceeds the 1980s threshold, and the last time total column abundances are below the threshold. Between the two time periods total column ozone values rise above and fall below the threshold.

Figure 4 shows the year raw total column ozone abundances are found to return to their 1980s values for the first time (light red) and final time (dark red) for each 10 degree latitude bin. In the tropics, total column ozone exceeds the 1980s threshold as early as 2000 which is due to the amplitude in total column ozone variations resulting from the solar cycle being greater than the trends due to ESC. However, despite this region seeing the first values greater than those of the 1980s, it is the only region in which total column ozone abundances are not greater than their 1980s values by the end of the simulation, consistent with other studies (e.g. Eyring et al., 2013; Meul et al., 2016). This is due to decreasing lower stratospheric ozone

15 concentrations resulting from an acceleration of the Brewer-Dobson circulation under increased greenhouse gas concentrations offsetting increased upper stratospheric ozone concentrations due to decreased ESC and increased CO<sub>2</sub> (explored in detail in Keeble et al., 2017).

In the northern hemisphere mid-latitudes earliest recovery occurs by  $\sim 2020$ , while final recovery occurs by 2040. The closeness of these two dates is due to the reduced interannual variability in the mid latitudes compared to both the tropics

20 and Arctic. The results are similar in the Southern hemisphere mid latitudes, although both dates are delayed by around 10 years, most likely due to the effects of Antarctic polar ozone depletion and transport of ozone poor airmasses into these latitudes upon the collapse of the Antarctic polar vortex.

Earliest recovery at high southern latitudes occurs by ~2040, with final recovery occurring by 2060. However, the signature of this recovery is very sensitive to month, and earlier signs of recovery may be identified in certain months (e.g. Solomon et

25 al., 2016). Arctic column ozone exhibits high interannual variability, with values exceeding the 1980s threshold as early as 2010. However, final recovery is not expected until ~2060.

The future evolution of total column ozone is dependent on the emissions scenario considered, and the exact timings of recovery will vary with changes to  $CO_2$ ,  $N_2O$  and  $CH_4$  emissions as well as ESC reductions. As a result, the expected return dates for each latitude will evolve as we approach those dates, in line with our increased understanding of the emissions activates or if future emission controls come into affect.

30 pathway or if future emission controls come into effect.

#### 7 Discussion and Conclusions

We have analysed total ozone values from an ensemble of UM-UKCA model runs to investigate different definitions of progress on the road to ozone recovery. This approach allows us to take into account the internal atmospheric variability of the model in assessing the statistical significance of each definition and so provide insight into similar analyses of

35 measurements. In particular, we have investigated three definitions: (i) a slowed rate of decline and the date of minimum ozone; (ii) the identification of significant positive trends; and (iii) a return to historic values.





The first and most obvious conclusion is that recovery can be identified in the first two metrics before a return to past thresholds is achieved. However, an important caveat is that while recovery may appear to be robust at a particular point of time, additional years of observations may lead to a reduced significance of trends due to natural variability (e.g. solar cycle, QBO, ENSO). This points to the need to ensure that the impact of natural cycles on total ozone is correctly described in the

- 5 statistical model. This is a challenge because of a number of factors. Firstly, the assumption in all MLR analysis of a linear relationship between the proxy variables used and the impact on ozone is not accurate, and there is growing evidence that these cycles are not isolated, but interact with one another (e.g. White and Liu, 2008; Calvo et al., 2009; Gray et al., 2010). Secondly, cycles with varying amplitudes (e.g. the solar cycle, which shows differing top of atmosphere solar flux during the last four solar maximums) or lengths (e.g. the QBO, the period of which may change in the future and has recently been
- 10 observed to undergo rapid, non-periodic reversal) have different impacts on total column ozone which makes accurate estimates of the coefficients for these variables in the MLR harder to achieve. Finally, volcanic eruptions are particularly difficult to account for in the MLR, both because of the infrequent, non-periodic timings of eruptions, and because eruptions have very different impacts on stratospheric ozone when stratospheric ESC concentrations are high compared to when ESC is low (e.g. Tie and Brasseur, 1995).
- 15 The mean trends of the ensemble members for 2000-2017 are positive at most latitudes. Unlike a recent analyses of total ozone measurements (Chipperfield et al., 2017), these trends are statistically significant. This increased significance results largely from the much larger sample size that arises in a multi-member ensemble and the resulting reduction in the uncertainty associated with the mean trend. The dates at which statistically significant trends are reached indicates that recovery of total column ozone began before 2017 at many latitudes in this ensemble of model runs.
- 20 The influence of the natural cycles on trend determination is least at latitudes where the trends are sizeable, as shown by the small differences between the year of first return and the year of final return as well as the negligible difference between the analysis using MLR analysis and that using raw data. For this reason, the mid-latitudes are the best place outside of Antarctica to identify recovery in the ozone column. This is due to the combination of reasonably large trend signal and comparably low variability (especially in the southern hemisphere). The Arctic is too variable for a signal to be easily
- 25 detected: this arises both from the large dynamic interannual variability and from the large changes in chemical ozone loss from year to year. The tropics have too small a trend compared to the natural variability resulting from the solar cycle and the QBO. This is most clearly seen in the large difference between the year at which recovery is first observed in the tropics and the year at which it remains above historic thresholds, though note that the latter year is influenced by the end of the model runs. While chemical ozone depletion is largest over Antarctica, the pronounced seasonal dependence of this
- 30 depletion, with the majority occurring during austral spring, means detection of trends is sensitive to which month is chosen. Solomon et al. (2016) highlight that while both September and October show high levels of chemical ozone depletion, the higher interannual variability in October compared to September (resulting from the break-up of the vortex in late Octoberearly November) means identification of ozone recovery is easier in September. As a result, Antarctica is a good location to look for early signs of recovery, but care must be taken when choosing which months to use, unlike in the mid-latitudes
- 35 where the seasonal cycle is relatively smaller.

Our analysis has been solely focused on interpreting the total ozone column record. Many studies have recently examined the trends in the vertical distribution of ozone since ESC maximized (e.g. Harris et al., 2015; Steinbrecht et al., 2017; Ball et al., 2017). In these studies, factors such as higher variability, greater uncertainties and poorer data quality add to the uncertainty in detection of significant trends compared to the total column. However similar studies to this one using

40 ensembles of model runs could provide real insights into the issue, especially in the climatically important lower stratosphere where ozone may still be decreasing (e.g. Ball et al., 2017).





# Data availability

Data from the two 1960-2100 transient simulations are available as part of the CCMI initiative through BADC: https://blogs.reading.ac.uk/ccmi/badc-data-access/. All further data are available upon request.

# **Competing interesting**

5 The authors declare that they have no conflict of interest.

## Acknowledgements

The research leading to these results has received funding from the European Community's Seventh Framework Programme (FP7/2007 - 2013) under grant agreement  $n^{\circ}$  603557 (StratoClim) and the European Research Council through the ACCI project (project number: 267760). We thank NCAS-CMS for modelling support. Model integrations have been performed

10 using the ARCHER UK National Supercomputing Service and MONSooN system, a collaborative facility supplied under the Joint Weather and Climate Research Programme, which is a strategic partnership between the UK Met Office and the Natural Environment Research Council.

# References

- 15 Austin, J., Hood, L. L., and Soukharev, B. E.: Solar cycle variations of stratospheric ozone and temperature in simulations of a coupled chemistry-climate model, Atmos. Chem. Phys., 7, 1693–1706, doi:10.5194/acp-7-1693-2007, 2007.
  - Avallone, L. M. and Prather, M. J.: Photochemical evolution of ozone in the lower tropical stratosphere, J. Geophys. Res., 101, 1457–1461, doi:10.1029/95JD03010, 1996.
- Baldwin, M. P., Gray, L. J., Dunkerton, T. J., Hamilton, K., Haynes, P. H., Randel, W. J., Holton, J. R., Alexander, M. J.,
  Hirota, I., Horinouchi, T., Jones, D. B. A., Kinnersley, J. S., Marquardt, C., Sato, K., and Takahashi, M.: The quasibiennial oscillation, Rev. Geophys., 39, 179–229, 2001.
  - Ball, W. T., Alsing, J., Mortlock, D. J., Staehelin, J., Haigh, J. D., Peter, T., Tummon, F., Stübi, R., Stenke, A., Anderson, J.,
    Bourassa, A., Davis, S. M., Degenstein, D., Frith, S., Froidevaux, L., Roth, C., Sofieva, V., Wang, R., Wild, J., Yu,
    P., Ziemke, J. R., and Rozanov, E. V.: Continuous decline in lower stratospheric ozone offsets ozone layer
- 25 recovery, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-862, in review, 2017.
  - Bednarz, E. M., Maycock, A. C., Abraham, N. L., Braesicke, P., Dessens, O., and Pyle, J. A.: Future Arctic ozone recovery: the importance of chemistry and dynamics, Atmos. Chem. Phys., 16, 12159-12176, doi:10.5194/acp-16-12159-2016, 2016.
- Bodeker, G. E., Shiona, H., and Eskes, H.: Indicators of Antarctic ozone depletion, Atmos. Chem. Phys., 5, 2603–2615,
  doi:10.5194/acp-5-2603-2005, 2005.
  - Braesicke, P., Keeble, J., Yang, X., Stiller, G., Kellmann, S., Abraham, N. L., Archibald, A., Telford, P., and Pyle, J. A.: Circulation anomalies in the Southern Hemisphere and ozone changes, Atmos. Chem. Phys., 13, 10677-10688, doi:10.5194/acp-13-10677-2013, 2013.
  - Brasseur, G.: The response of the middle atmosphere to long-term and short-term solar variability: A two-dimensional model, J. Geophys. Res., 98, 23079–23090, 1993.





5

- Calisesi, Y. and Matthes, K.: The Middle Atmospheric Ozone Response to the 11-Year Solar Cycle, Springer New York, 23, 273-286, doi:10.1007/978-0-387-48341-2\_22, 2007.
- Calvo, N., Giorgetta, M. A., Garcia-Herrera, R., and Manzini, E.: Nonlinearity of the combined warm ENSO and QBO effects on the Northern Hemisphere polar vortex in MAECHAM5 simulations, J. Geophys. Res.-Atmos., 114, D20, doi:10.1029/2008JD011445, 2009.

- Chehade, W., Weber, M., and Burrows, J. P.: Total ozone trends and variability during 1979-2012 from merged data sets of various satellites, Atmos. Chem. Phys., 14, 7059-7074, https://doi.org/10.5194/acp-14-7059-2014, 2014.
- Chipperfield, M. P. and Feng, W.: Comment on: Stratospheric Ozone Depletion at northern mid-latitudes in the 21st century: The importance of future concentrations of greenhouse gases nitrous oxide and methane, Geophys. Res. Lett., 30, 1389, doi:10.1029/2002GL016353, 2003.
- Chipperfield, M., Liang, Q., Strahan, S., Morgenstern, O., Dhomse, S., Abraham, N., Archibald, A., Bekki, S., Braesicke, P., Di Genova, G., Fleming, E. L., Hardiman, S. C., Iachetti, D., Jackman, C. H., Kinnison, D. E., Marchand, M., Pitari, G., Pyle, J. A., Rozanov, E., Stenke, A., and Tummon, F.: Multimodel estimates of atmospheric lifetimes of long-lived ozone-depleting substances: Present and future, J. Geophys. Res. Atmos., 119, 2555-2573, 2014.
- 15 Chipperfield, M., Bekki, S., Dhomse, S., Harris, N., Hassler, B., Hossaini, R., Steinbrecht, W., Thiéblemont, R., and Weber, M.: Detecting recovery of the stratospheric ozone layer, Nature, doi:10.1038/nature23681, 2017.
  - Eyring, V., Waugh, D. W., Bodeker, G. E., Cordero, E., Akiyoshi, H., Austin, J., Beagley, S. R., Boville, B., Braesicke, P., Bruhl, C., Butchart, N., Chipperfield, M. P., Dameris, M., Deckert, R., Deushi, M., Frith, S. M., Garcia, R. R., Gettelman, A., Giorgetta, M., Kinnison, D. E., Mancini, E., Manzini, E., Marsh, D. R., Matthes, S., Nagashima T.,
- 20 Newman, P. A., Nielsen, J. E., Pawson, S., Pitari, G., Plummer, D. A., Rozanov, E., Schraner, M., Scinocca, J. F., Semeniuk K., Shepherd, T. G., Shibata, K., Steil, B., Stolarski, R., Tian, W., and Yoshiki, M.: Multimodel projections of stratospheric ozone in the 21st century, J. Geophys. Res., 112, D16303, doi:10.1029/2006JD008332, 2007.
- Eyring, V., Chipperfield, M. P., Giorgetta, M. A., Kinnison, D. E., Manzini, E., Matthes, K., Newman, P. A., Pawson, S., 25 Shepherd, T. G., and Waugh, D. W.: Overview of the New CCMVal Reference and Sensitivity Simulations in Support of Upcoming Ozone and Climate Assessments and the Planned SPARC CCMVal Report, SPARC Newsletter No. 30, 20-26, 2008.
  - Eyring, V., Cionni, I., Bodeker, G. E., Charlton-Perez, A. J., Kinnison, D. E., Scinocca, J. F., Waugh, D. W., Akiyoshi, H., Bekki, S., Chipperfield, M. P., Dameris, M., Dhomse, S., Frith, S. M., Garny, H., Gettelman, A., Kubin, A.,
- 30 Langematz, U., Mancini, E., Marchand, M., Nakamura, T., Oman, L. D., Pawson, S., Pitari, G., Plummer, D. A., Rozanov, E., Shepherd, T. G., Shibata, K., Tian, W., Braesicke, P., Hardiman, S. C., Lamarque, J. F., Morgenstern, O., Pyle, J. A., Smale, D., and Yamashita, Y.: Multi-model assessment of stratospheric ozone return dates and ozone recovery in CCMVal-2 models, Atmos. Chem. Phys., 10, 9451-9472, https://doi.org/10.5194/acp-10-9451-2010, 2010.
- 35 Eyring, V., Arblaster, J. M., Cionni, I., Sedláček, J., Perlwitz, J., Young, P. J., Bekki, S., Bergmann, D., Cameron-Smith, P., Collins, W. J., Faluvegi, G., Gottschaldt, K.-D., Horowitz, L. W., Kinnison, D. E., Lamarque, J.-F., Marsh, D. R., Saint-Martin, D., Shindell, D. T., Sudo, K., Szopa, S., and Watanabe, S.: Long-term ozone changes and associated climate impacts in CMIP5 simulations, J. Geophys. Res.-Atmos., 118, 5029-5060, doi:10.1002/jgrd.50316, 2013.
- Gray, L. J., Beer, J., Geller, M., Haigh, J. D., Lockwood, M., Matthes, K., Cubasch, U., Fleitmann, D., Harrison, G., Hood, 40 L., Luterbacher, J., Meehl, G. A., Shindell, D., van Geel, B., and White, W.: Solar Influences on Climate, Rev. Geophys., 48, G4001, doi:10.1029/2009RG000282, 2010.





- Haigh, J. D. and Pyle, J. A.: Ozone perturbation experiments in a two-dimensional circulation model, Q. J. Roy. Meteor. Soc., 108, 551–574, doi:10.1002/qi.49710845705, 1982.
- Harris, N. R. P., Kyro, E., Staehelin, J., Brunner, D., Andersen, S.-B., Godin-Beekmann, S., Dhomse, S., Hadjinicolaou, P., Hansen, G., Isaksen, I., Jrrar, A., Karpetchko, A., Kivi, R., Knudsen, B., Krizan, P., Lastovicka, J., Maeder, J.,
- 5

Hansen, G., Isaksen, I., Jirar, A., Karpetchko, A., Kivi, K., Khudsen, B., Krizan, P., Lastovicka, J., Maeder, J., Orsolini, Y., Pyle, J. A., Rex, M., Vanicek, K., Weber, M., Wohltmann, I., Zanis, P., and Zerefos, C.: Ozone trends at northern mid- and high latitudes – a European perspective, Ann. Geophys., 26, 1207–1220, doi:10.5194/angeo-26-1207-2008, 2008.

Harris, N. R. P., Hassler, B., Tummon, F., Bodeker, G. E., Hubert, D., Petropavlovskikh, I., Steinbrecht, W., Anderson, J., Bhartia, P. K., Boone, C. D., Bourassa, A., Davis, S. M., Degenstein, D., Delcloo, A., Frith, S. M., Froidevaux, L.,

- Godin-Beekmann, S., Jones, N., Kurylo, M. J., Kyrölä, E., Laine, M., Leblanc, S. T., Lambert, J.-C., Liley, B., Mahieu, E., Maycock, A., de Mazière, M., Parrish, A., Querel, R., Rosenlof, K. H., Roth, C., Sioris, C., Staehelin, J., Stolarski, R. S., Stübi, R., Tamminen, J., Vigouroux, C., Walker, K. A., Wang, H. J., Wild, J., and Zawodny, J. M.: Past changes in the vertical distribution of ozone Part 3: Analysis and interpretation of trends, Atmos. Chem. Phys., 15, 9965-9982, doi:10.5194/acp-15-9965-2015, 2015.
- 15 Hewitt, H. T., Copsey, D., Culverwell, I. D., Harris, C. M., Hill, R. S. R., Keen, A. B., McLaren, A. J., and Hunke, E. C.: Design and implementation of the infrastructure of HadGEM3: the next-generation Met Office climate modelling system, Geosci. Model Dev., 4, 223-253, doi:10.5194/gmd-4-223-2011, 2011.
  - Hollandsworth, S. M., Bowman, K. P., and McPeters, R. D.: Observational study of the quasi-biennial oscillation in ozone, J. Geophys. Res.-Atmos., 100, 7347–7361, doi:10.1029/95JD00193, 1995.
- 20 Iglesias-Suarez, F., Young, P. J., and Wild, O.: Stratospheric ozone change and related climate impacts over 1850–2100 as modelled by the ACCMIP ensemble, Atmos. Chem. Phys., 16, 343-363, doi:10.5194/acp-16-343-2016, 2016.
  - Jones, A., Urban, J., Murtagh, D. P., Eriksson, P., Brohede, S., Haley, C., Degenstein, D., Bourassa, A., von Savigny, C., Sonkaew, T., Rozanov, A., Bovensmann, H., and Burrows, J.: Evolution of stratospheric ozone and water vapour time series studied with satellite measurements, Atmos. Chem. Phys., 9, 6055–6075, doi:10.5194/acp-9-6055-2009, 2009.
- 25

- Keeble, J., Braesicke, P., Abraham, N. L., Roscoe, H. K., and Pyle, J. A.: The impact of polar stratospheric ozone loss on Southern Hemisphere stratospheric circulation and climate, Atmos. Chem. Phys., 14, 13705-13717, doi:10.5194/acp-14-13705-2014, 2014.
- Keeble, J., Bednarz, E. M., Banerjee, A., Abraham, N. L., Harris, N. R. P., Maycock, A. C., and Pyle, J. A.: Diagnosing the
   radiative and chemical contributions to future changes in tropical column ozone with the UM-UKCA chemistry– climate model, Atmos. Chem. Phys., 17, 13801-13818, https://doi.org/10.5194/acp-17-13801-2017, 2017.
  - Leblanc, T. and McDermid, I. S.: Quasi-biennial oscillation signatures in ozone and temperature observed by lidar at Mauna Loa, Hawaii (19.5°N, 155.6°W), J. Geophys. Res.-Atmos., 106, 14869–14874, doi:10.1029/2001JD900162, 2001.
  - Lee, H. and Smith, A.: Simulation of the combined effects of solar cycle, quasi-biennial oscillation, and volcanic forcing on
  - stratospheric ozone changes in recent decades, J. Geophys. Res.-Atmos., 108, D2, doi:10.1029/2001JD001503, 2003.
  - Mäder, J. A., Staehelin, J., Peter, T., Brunner, D., Rieder, H. E., and Stahel, W. A.: Evidence for the effectiveness of the Montreal Protocol to protect the ozone layer, Atmos. Chem. Phys., 10, 12161-12171, doi:10.5194/acp-10-12161-2010, 2010.
- 40 Manzini, E.: Atmospheric science: ENSO and the stratosphere, Nat. Geosci., 2, 749–750, 2009.





5

- McLandress, C., Shepherd, T. G., Scinocca, J. F., Plummer, D. A., Sigmond, M., Jonsson, A. I., and Reader, M. C.: Separating the dynamical effects of climate change and ozone depletion, Part II: Southern Hemisphere troposphere, J. Climate, 24, 1850–1868, 2011.
- Meul, S., Langematz, U., Oberländer, S., Garny, H., and Jöckel, P.: Chemical contribution to future tropical ozone change in the lower stratosphere, Atmos. Chem. Phys., 14, 2959–2971, doi:10.5194/acp-14-2959-2014, 2014.
- Meul, S., Dameris, M., Langematz, U., Abalichin, J., Kerschbaumer, A., Kubin, A., and Oberländer-Hayn, S.: Impact of rising greenhouse gas concentrations on future tropical ozone and UV exposure, Geophys. Res. Lett., 43, 2919– 2927, https://doi.org/10.1002/2016GL067997, 2016.
- Molina, M. J. and Rowland, F. S.: Stratospheric sink for chlorofluoromethanes: chlorine atomcatalysed destruction of ozone, Nature, 249, 810–812, doi: 10.1038/249810a0, 1974.
- Morgenstern, O., Braesicke, P., O'Connor, F. M., Bushell, A. C., Johnson, C. E., Osprey, S. M., and Pyle, J. A.: Evaluation of the new UKCA climate-composition model – Part 1: The stratosphere, Geosci. Model Dev., 2, 43-57, doi:10.5194/gmd-2-43-2009, 2009.
- Morgenstern, O., Hegglin, M. I., Rozanov, E., O'Connor, F. M., Abraham, N. L., Akiyoshi, H., Archibald, A. T., Bekki, S.,
  Butchart, N., Chipperfield, M. P., Deushi, M., Dhomse, S. S., Garcia, R. R., Hardiman, S. C., Horowitz, L. W.,
  Jöckel, P., Josse, B., Kinnison, D., Lin, M., Mancini, E., Manyin, M. E., Marchand, M., Marécal, V., Michou, M.,
  Oman, L. D., Pitari, G., Plummer, D. A., Revell, L. E., Saint-Martin, D., Schofield, R., Stenke, A., Stone, K., Sudo,
  K., Tanaka, T. Y., Tilmes, S., Yamashita, Y., Yoshida, K., and Zeng, G.: Review of the global models used within
  phase 1 of the Chemistry–Climate Model Initiative (CCMI), Geosci. Model Dev., 10, 639-671,
  https://doi.org/10.5194/gmd-10-639-2017, 2017.
  - Nair, P. J., Godin-Beekmann, S., Kuttippurath, J., Ancellet, G., Goutail, F., Pazmiño, A., Froidevaux, L., Zawodny, J. M., Evans, R. D., Wang, H. J., Anderson, J., and Pastel, M.: Ozone trends derived from the total column and vertical profiles at a northern mid-latitude station, Atmos. Chem. Phys., 13, 10373–10384, https://doi.org/10.5194/acp-13-10373-2013, 2013.
- 25 Newchurch, M. J., Yang, E.-S., Cunnold, D. M., Reinsel, G. C., Zawodny, J. M., and Russell, J. M.: Evidence for slowdown in stratospheric ozone loss: first stage of ozone recovery, J. Geophys. Res., 108, 4507, https://doi.org/10.1029/2003JD003471, 2003.
  - Newman, P. A., Nash, E. R., Kawa, S. R., Montzka, S. A., and Schauffler, S. M.: When will the Antarctic ozone hole recover?, Geophys. Res. Lett., 33, L12814, doi:10.1029/2005GL025232, 2006.
- 30 Plumb, R. A.: A 'tropical pipe' model of stratospheric transport, J. Geophys. Res., 101, 3957–3972, 1996.
  - Randel, W. J. and Wu, F.: TOMS total ozone trends in potential vorticity coordinates, Geophys. Res. Lett., 22, 683–686, 1995.
    - Randel, W. J., Garcia, R. R., Calvo, N., and Marsh, D.: ENSO influence on zonal mean temperature and ozone in the tropical lower stratosphere, Geophys. Res. Lett., 36, L15822, doi:10.1029/2009GL039343, 2009.
- 35 Ravishankara, A. R., Daniel, J. S., and Portmann, R. W.: Nitrous Oxide (N<sub>2</sub>O): The Dominant Ozone Depleting Substance Emitted in the 21st Century, Science, 326(123), 682-125, 2009.
  - Reinsel, G. C., Miller, A. J., Weatherhead, E. C., Flynn, L. E., Nagatani, R. M., Tiao, G. C., and Wuebbles, D. J.: Trend analysis of total ozone data for turnaround and dynamical contributions, J. Geophys. Res., 110, D16306, https://doi.org/10.1029/2004JD004662, 2005.
- 40 Revell, L. E., Bodeker, G. E., Huck, P. E., Williamson, B. E., and Rozanov, E.: The sensitivity of stratospheric ozone changes through the 21st century to N<sub>2</sub>O and CH<sub>4</sub>, Atmos. Chem. Phys., 12, 11309–11317, doi:10.5194/acp-12-11309-2012, 2012.





Rowland, F. S. and Molina, M. J.: Chlorofluoromethanes in environment, Rev. Geophys., 13, 1-35, 1975.

Solomon, S., Ivy, D. J., Kinnison, D., Mills, M. J., Neely, R. R., and Schmidt, A.: Emergence of healing in the Antarctic ozone layer, Science, 353, 269-274, doi:10.1126/science.aae0061, 2016.

SPARC: SPARC assessment of stratospheric aerosol properties (ASAP), Tech. Rep. WMO-TD No. 1295, WCRP Series Report No. 124, SPARC Report No. 4, Berrieres le Buisson, Cedex, 2006.

SPARC: SPARC Report on the Lifetimes of Stratospheric Ozone-Depleting Substances, Their Replacements, and Related Species, Edited by M. K. W. Ko, P. A. Newman, S. Reimann, and S. E. Strahan, SPARC Report No. 6, WCRP-15/2013, 2013.

Staehelin, J., Harris, N. R. P., Appenzeller, C., Eberhard, J., and Piechowski, M.: Observations of ozone trends, Rev. Geophys., 39, 231-290, 2001.

Steinbrecht, W., Froidevaux, L., Fuller, R., Wang, R., Anderson, J., Roth, C., Bourassa, A., Degenstein, D., Damadeo, R., Zawodny, J., Frith, S., McPeters, R., Bhartia, P., Wild, J., Long, C., Davis, S., Rosenlof, K., Sofieva, V., Walker, K., Rahpoe, N., Rozanov, A., Weber, M., Laeng, A., von Clarmann, T., Stiller, G., Kramarova, N., Godin-Beekmann, S., Leblanc, T., Querel, R., Swart, D., Boyd, I., Hocke, K., Kämpfer, N., Maillard Barras, E., Moreira,

15 L., Nedoluha, G., Vigouroux, C., Blumenstock, T., Schneider, M., García, O., Jones, N., Mahieu, E., Smale, D., Kotkamp, M., Robinson, J., Petropavlovskikh, I., Harris, N., Hassler, B., Hubert, D., and Tummon, F.: An update on ozone profile trends for the period 2000 to 2016, Atmos. Chem. Phys., 17, 10675-10690, https://doi.org/10.5194/acp-17-10675-2017, 2017. Stolarski, R. S., and Cicerone, R. J.: Stratospheric Chlorine -Possible Sink for Ozone, Canadian Journal of Chemistry- Revue Canadienne De Chimie, 52, 1610-1615, Doi 10.1139/V74-233, 1974.

- Telford, P. J., Braesicke, P., Morgenstern, O., and Pyle, J.: Reassessment of causes of ozone column variability following the eruption of Mount Pinatubo using a nudged CCM, Atmos. Chem. Phys., 9, 4251-4260, doi:10.5194/acp-9-4251-2009, 2009.
- Tie, X., and Brasseur, G.: The response of stratospheric ozone to volcanic eruptions: sensitivity to atmospheric chlorine loading, Geophys. Res. Lett., 22, 3035-3038, 1995.
- van Loon, H. and Labitzke, K.: The influence of the 11-year solar cycle on the stratosphere below 30 km: A review, Space Sci. Rev., 94, 259-278, 2000.

Weatherhead, E. C and Andersen, S. B.: The search for signs of recovery of the ozone layer, Nature, 441, 39-45, 2006.

- White, W. B. and Liu, Z.: Non-linear alignment of El Niño to the 11-yr solar cycle, Geophys. Res. Lett., 35, L19607, doi:10.1029/2008GL034831, 2008
- World Meterological Organization (WMO): Scientific Assessment of Ozone Depletion: 2006, Global Ozone Research and Monitoring Project, Report No. 50, Geneva, Switzerland, 2007.
- World Meterological Organization (WMO): Scientific Assessment of Ozone Depletion: 2010, Global Ozone Research and Monitoring Project, Report No. 52, Geneva, Switzerland, 2011.
- 35 World Meterological Organization (WMO): Scientific Assessment of Ozone Depletion: 2014, Global Ozone Research and Monitoring Project, Report No. 55, Geneva, Switzerland, 2014.

10

5







**Figure 1:** Deseasonalised total column ozone anomalies (in DU) relative to the 1980 mean, averaged over 60°S-60°N, for the seven UM-UKCA transient ensemble members (light blue lines) and ensemble mean (dark blue line). Also shown are the ozone residuals calculated when natural cycles are removed from each ensemble member (light red lines) and the mean of the ozone residuals (dark red line). The inset shows total column ozone anomalies for the transient UM-UKCA

<sup>5</sup> of the ozone residuals (dark red line). The inset shows total column ozone anomalies for the transient simulations and v2.8 of the Bodeker dataset (Bodeker et al., 2005; black line) from 1975 to 2015.







Figure 2: Ozone trends from 1980-1997 (blue points) and 2000-2017 (red points) for the raw modelled data (dark points) and MLR residuals (light points). Error bars associated with each trend represent the 95% confidence intervals.





5



**Figure 3:** Year when recovery trend becomes significant, where trends are defined as significantly different from zero when they have a p-value < 0.05. Trend significance is determined for the ozone residuals calculated using the MLR coefficients, and a distinction is made for the first time significance can be determined (blue points) and the time after which trends remain significant (red points). Error bars represent the 95% confidence intervals.





5



**Figure 4:** Year total column ozone returns to 1980 annual mean values in each latitude band for the raw data from the seven UM-UKCA ensemble members. Blue points represent the first time annual mean values exceed the 1980 mean, while red points represent the final time annual mean values are lower than the 1980 mean. Error bars represent the 95% confidence intervals.